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The Atomic Industry and Human Ecology

I—Waste Disposal

THERE IS STILL TIME to ensure that waste from the atomic industry never becomes unacceptably hazardous or a nuisance. This result requires the best efforts of intelligent and responsible men who are abundantly supplied with authority and equipment. These efforts must be continuous and kept free even from suspicion of secret influence. Greater efforts are needed than are now being made, and more will be suddenly and gravely needed in the event of war.

By waste disposal we mean simply, "do what one will with, regulate, get off one's hands, stow away, settle, finish, kill, demolish, dismiss, disperse, decay, consume, etc.," the various products that are unused by the producer in the ultimate (if not immediate) interest of the consumer. It is sometimes said that modern meat-packing establishments "waste" nothing but the squeal of a pig. This is no doubt true, but direct radiation or stack effluent from a nuclear reactor, for example, requires more care in its disposal than the odors and aerosols from industry generally have received.

Waste disposal presents problems fundamental to all living things and when, as usually occurs, the adopted disposal procedures change environment, then the matter is necessarily included in any discussion of ecology. Man is by no means the most orderly of creatures, either at home or at work, and the refuse of the suburb or of any other small town is not unlike that of the largest city. Men are frequently in disagreement on waste disposal, even in principle.

These are fundamental problems and deserve the consideration of thinking men. They need the attention even of the social scientists from whom mankind expects leadership in how education may replace or enforce the Golden Rule. And scientific research and engineering applications of the highest order are required to determine effective and efficient disposal methods. Finally public health authorities and their several specialized methods and personnel must educate, interpret, and guarantee that acceptable results are obtained.

The differences and similarities between waste products from the atomic and the chemical industries, say, need not be labored. It is sufficient at this point to recall that conditions hazardous to health may result from the atomic industry without being determinable by any of the five senses. This fact will be of advantage in clarifying issues which may arise between those responsible for the source and those objecting to it, for one may anticipate (aside from action taken by cranks) litigation only when instrumental records show a radiation count, for example, that has been declared improper by responsible authority.

Proper operating levels of various sources are presently under study. There is not complete unanimity on what amounts of radiation may be "harmful." For example, a recent survey of opinion among radiologists showed estimates ranging from 100 to 1,000 roentgens for the near-lethal human dose of radiation in a two-hour period of total immersion. The question whether, in fact, this or that dose will kill is much more difficult for the peacetime health physicist to answer in practice than is the comparable question for his counterpart in war—namely, how many of a large number will die if exposed to a particular radiation intensity, over all or a part of the body, for a certain interval of time. The question becomes further complicated since no two individuals will have the same radiation history. In any case radiation levels ought surely to be kept as low as suggested by the experience acquired and, *always and carefully*, measured with the best available instruments.

Persons placed in positions of responsibility with respect to the hazards of radioactive wastes, or other wastes such as beryllium dust, have reacted similarly to their responsibilities, when the hazard has been recognized as such. They have taken the safe course to the extent that the accident record of the Manhattan District and of the Atomic Energy Commission and its contractors has been superior to that of any other industry with a comparable number of persons engaged in comparable work. The public has not been exposed to hazard at all so far as is known publicly. The industry is not complacent even among those who use small quantities of radioactive material and, usually, dump their waste down the drain or blow it out of a stack.

One must consider a number of factors in evaluating the hazards of any new work of man. Such factors as the expected good are balanced against the possible loss. One takes a calculated risk. It is known in advance, for example, that any public work of consequence will cost a few lives, whether the project be a bridge, dam, tall building, etc. The individual engaged on the project accepts his chances of survival, so long as his estate is protected financially, with no more hesitation than in crossing a busy street against traffic. Insurance firms evaluate these matters statistically and establish premiums to cover the risk. Atomic workers are now, almost without exception, covered for the same premiums paid by those outside their industry; and no area outside any atomic plant's domain requires an increased premium for insurance against radiation hazard.

The central question in consideration of waste disposal in the atomic industry, from the ecological viewpoint, is whether the public will require complete control over all effluent, at least to the point where, by measurement and observation, succeeding generations may learn its complete effects on man. There is still time to determine the intensity of background radiation from natural sources in most areas. Whether the background can be increased two-, three-, or N-fold without unpleasant results is unknown. Some practical limit must obviously be set by general agreement. It is unthinkable that man will not proceed to develop the atomic industry fully; it is unacceptable that this or any succeeding generation will so change man's environment that man cannot survive.

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Control room of British reactor "Gleep" (graphite low energy experimental pile)

Instrumentation for a Nuclear Reactor*

By D. COCHRAN and C. A. HANSEN, Jr.

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MANY UNUSUAL PROBLEMS complicate the application of instrumentation in an atomic power plant. Some of these arise from the presence of intense radiation and high temperatures in much of the system. As yet there are not demonstrated solutions to all these problems.

The Atomic Power Plant

The atomic power plant now being constructed for the Atomic Energy Commission at the Knolls Atomic Power Laboratory in Schenectady will probably be the first to generate useful electric power in more than token amounts. This is an experimental plant whose purpose is to investigate power production on a practical scale. The possibility of *breeding*, which is defined as the simultaneous production of new nuclear fuel at a rate greater than the fuel consumption, will also be investigated. This plant employs liquid metal reactor coolant and steam turbo-electric power generation equipment.

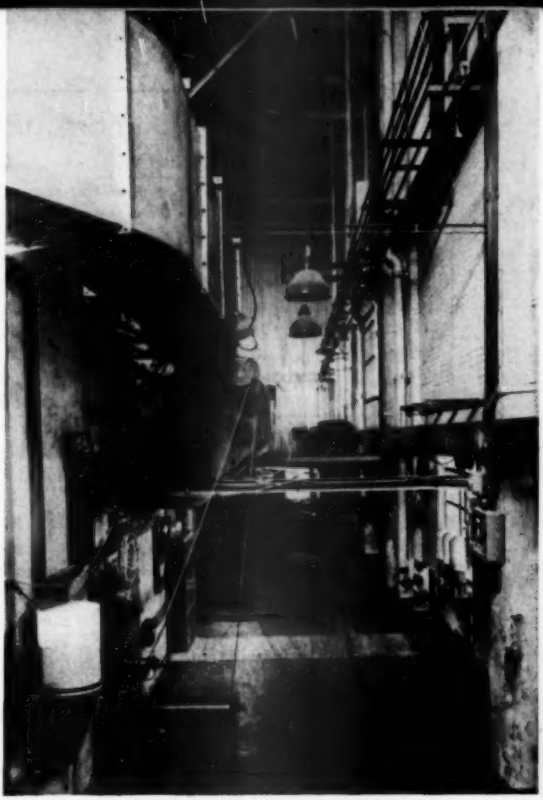
The reactor is an assembly consisting

* From a talk delivered June 28 at the semi-annual meeting of the American Society of Mechanical Engineers in San Francisco.

† Operated for the Atomic Energy Commission by the General Electric Research Laboratory.

of jacketed nuclear fuel elements, control elements, moderating material and reflecting material, all supported and positioned in a structural matrix. The liquid metal coolant is pumped through channels in the reactor core, where it receives heat from the fuel elements. Leaving the reactor the coolant passes through heat exchangers where it gives up heat to evaporate and superheat steam. The steam drives a turbine to generate electric power.

The heat-generating nuclear reaction in the reactor core causes tremendous radioactivity which must be confined by an absorbing shield. The liquid metal coolant passing through the reactor core becomes radioactive because of its exposure to neutron flux. Thus, the piping, pumps, heat exchangers and storage tanks which carry the radioactive coolant also must be shielded, although to a lesser extent than the reactor core. The radioactivity of the coolant does not cause radioactivity in the water and steam passing through the heat exchangers; hence, it is not necessary to shield the stream and water piping and equipment in the power plant.



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Control face of "Gleep" is shown on the left. In the bottom corner can be seen the chambers containing boron trifluoride gas which are used to measure the intensity of neutrons in the pile, and hence to control the power level at which the pile is operated. On the right are some health monitoring instruments and behind them is the control room

The reactor coolant leaves the reactor at temperatures high enough to permit an efficient steam cycle. Liquid metals have relatively low vapor pressures, however, and can be maintained in the liquid phase at pressures of a few atmospheres throughout the circuit. Hot liquid metals oxidize readily in contact with air, forming compounds harmful to operation as a coolant. To prevent oxidation it is necessary to maintain a blanket of inert gas over free surfaces of liquid metal wherever they occur.

There are remotely controlled mechanisms for actuating control elements, for removing spent fuel elements and for inserting fresh ones. The spent fuel elements are intensely radioactive, and must be transported, stored and processed in shielded spaces, by means of remotely controlled machinery.

Operation of this plant requires much elaborate instrumentation. Most of

PLANT OPERATION

Quantities to Be Detected	*Range*
1. Neutron flux density in or near the reactor core	10^7 fold
2. Control element position	Within 0.005" over many feet
3. Reactor coolant temperatures	200° F to 1200° F
4. Reactor coolant pressures	−10 psi to 100 psi
5. Reactor coolant flow	0 to thousands of gpm
6. Coolant liquid level	
7. Blanket gas temperature	200° F to 1200° F
8. Blanket gas pressure	1 mm Hg abs to 100 psi

the instruments are standard commercial devices, as, for example, those on the steam turbine, electric generating equipment and auxiliaries. Others must meet unusual requirements, and special designs are required. These can be classified according to the general function they perform as follows:

1. Operation. Instruments required for the control and safety of the plant operation.

2. Maintenance. Instruments to observe the condition and behavior of the reactor structure, fuel elements, coolant and blanketing gas as the operation of the plant proceeds.

3. Health. Instruments required to monitor radiation leakage in the plant to insure protection of operating personnel.

For plant *operation* there must be instruments to detect the quantities shown in the table above.

To assist and guide *maintenance*, the quantities shown in the table on page 7 must be detected or observed.

Health instruments must detect alpha, beta, gamma, and neutron radiation in exceedingly minute quantities, wherever it may leak through shielding, emanate from radioactive objects or from surfaces contaminated by radioactive substance. There is considerable published information about these instruments, so this article will not elaborate further on them.*

General Design Requirements

In addition to requisite accuracy and sensitivity, a number of detecting devices in atomic power plants must meet requirements not found in conventional power plant applications. These requirements arise from the radioactivity of the reactor and coolant and the shielding surrounding them. The structure, equipment and instruments within this radioactive zone are subjected to radiation of varying intensity, depending on location. Some of these become permanently radioactive as a result of this exposure.

Following are some of the unusual requirements of instrumentation in the radioactive zone:

1. Extreme dependability. It is essential that the nuclear reaction remain under control at all times. This is necessary to protect the reactor and associated equipment—a major investment—from damage. It is also necessary to protect the employees in the power plant from exposure to radiation from the radioactive materials in the reactor. It is important here to note that there is absolutely no way in which a power nuclear reactor can

* L. L. German, H. M. Rosendaal, *Elec. Eng.* 67, 884 (1948).

explode like an atomic bomb. A power nuclear reactor differs radically in basic construction from an atomic bomb and just cannot be made to explode like a bomb.

It is still very important to provide ultra-reliable operating controls backed up by pyramided safety controls. Safety controls must be designed to "put out the fire," not to "blow off steam" as can be done by safety valves on a conventional steam boiler. There is nothing about a nuclear reactor that corresponds to the safety valve of a steam boiler in a conventional power plant. In such a plant when there is a sudden increase in the rate of heat generation, or, as is more likely, a sudden decrease in the rate of heat absorption by the turbine system, the extra energy in the boiler may be dissipated by blowing the safety valve. The nuclear reactor, however, has a definitely limited capacity for absorbing or dissipating extra energy. The operating controls, backed up by safety controls, must be able to decrease the rate of heat generation quickly enough to prevent overheating of the reactor even under the most abnormal fluctuations in connected load or failure of power conversion equipment.

Of course, no single instrument bears the entire responsibility for the safety of the plant. There are a multiplicity of detecting devices and a number of independent safety controls. Since an exceedingly high order of safety is to be achieved, these many instruments and controls are designed to "fail safe." So, an instrument failure shuts down the plant. Uninterrupted operation of the plant, therefore, requires reliability of all instruments and circuits.

PLANT MAINTENANCE

Quantities to Be Detected or Observed	*Range*
1. Fuel element temperatures	200° F to 1200° F
2. Internal pressures in reactor parts	A few atmospheres
3. Fuel element rupture	Traces of fission products and fuel released into coolant stream
4. Core structure and fuel element distortions	A few mils
5. Chemical contamination of reactor coolant	Very small amounts
6. Fission product (radioactive) contamination of coolant	Traces of neutron flux in coolant remote from reactor
7. Chemical contamination of blanket gas	Very small amounts
8. Fission product contamination of blanket gas	Traces of radioactivity in gas withdrawn from radioactive zone
9. Fuel handling manipulation	Remote visual inspection of motions of a few hundredths of an inch to several feet
10. Location of stored spent fuel-elements	A fraction of an inch in channels and passageways several hundred feet long
11. Leakage of coolant	1 cc and up

Fission products emitting high levels of radiation are processed inside a thick-walled concrete cell at Oak Ridge National Laboratory. The high intensity radiation present in such operations creates problems similar to those with reactors. Photo shows apparatus on outside wall where all operations are performed by remote control. The radiation emitted through an opening is being checked

2. Remote maintenance. It is not possible to perform direct inspection and maintenance work on the detecting devices within the radioactive zone. They must be designed so that they can be checked and calibrated remotely, where this is necessary.

When a failure occurs, it may be necessary, because of radioactivity at the location of the device, to remove and replace it by means of remotely operated equipment. If the device has become radioactive in service it usually cannot be repaired, after removal, but must be replaced with a new unit.

Where the detecting elements are very difficult to remove or are completely inaccessible, stand-by detecting elements must be installed at the time of erection.

3. Neutron and gamma radiation. Pressure, temperature and flow detectors must function correctly in the presence of tremendous radiation. Continued exposure to radiation must not cause such physical deterioration of the device as to change its performance in a way that cannot be determined and compensated for.

Devices to detect chemical contamination of the coolant must function in spite of the radioactivity of the coolant.

4. Absolute leak tightness. Detecting elements exposed to the coolant or blanket gas must be absolutely leak-tight. The escape of even minute quantities of radioactive coolant or of blanket gas which might contain gaseous fission products would cause radiation monitoring instruments to register requiring that the plant be shut down and evacuated until the leak could be located and repaired.

5. Long service. Because of the extreme difficulty and expense of replacing faulty detecting elements, long operating life is much more important in these applications than is usually the

case. There is great incentive to develop long-lived devices, and there is frequent justification for using more expensive devices to insure long service. A few situations exist where, once the reactor has been operated, detecting elements become absolutely inaccessible. Such arrangements are obviously undesirable, and much effort must be devoted to designing systems that avoid them.

Instrument Applications

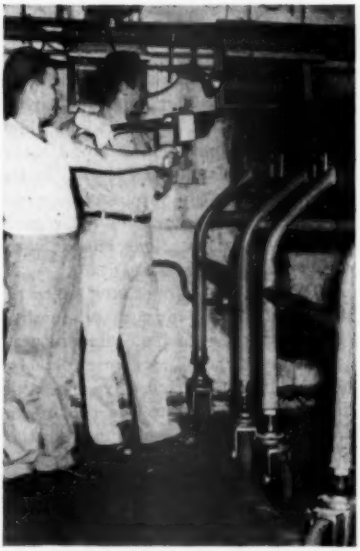
As stated at the beginning of this article, as yet there are not successfully demonstrated devices to meet these requirements for all the applications listed. Some of the devices being worked on are discussed here.

1. Neutron flux. Neutrons can be detected by measuring alpha radiation induced by their capture in boron. This is done by means of ion chambers coated with boron or filled with boron-trifluoride gas. Boron atoms present a relatively large target for neutrons, and upon capture, emit alpha particles which are powerfully ionizing in gas. Thus, the capture of neutrons causes ion current pulses between electrodes of the ion chamber which are readily discriminated from ionization caused by gamma radiation.

This discrimination is necessary because power generation in the reactor is directly related to neutron flux density, but not to the level of gamma radiation.

In operating the reactor it is necessary to detect small changes in neutron flux over a range of 10^6 or 10^7 in intensity. Ion currents in the order of micro-micro amperes must be measured. To extend the range, several ion chambers may be used having different ranges by virtue of their location with respect to the reactor core.

A further requirement of the neutron detecting systems is that they have an extremely rapid response time, in the order of milliseconds. This speed is necessary to actuate control elements or safety elements to compensate for sudden changes in reactivity in the reactor.



Heavy lead shields in the foreground receive the hot materials withdrawn from the reactor. The radiation must be measured to be sure that safety limits are not exceeded

The principal limitations as to range and speed of neutron detection lie not in the ion chambers but in the associated circuits and relays. Present high sensitivity amplifiers have high input impedance which imposes difficulties as to electrically insulating and shielding the leads to the detector so as to exclude spurious electric signals.

Finally, the neutron detectors must operate accurately and reliably at the high temperatures prevailing around the reactor core. This is a new requirement not found in the neutron detectors used in health physics work or in controlling low temperature reactors, such as those at Hanford, which do not generate useful power.

2. Pressure. Pressure of the reactor coolant can be measured by bourdon-type pressure gages with electrical telemetering. A gage fluid must be selected which will not suffer deterioration because of radiation exposure, and which will not contaminate the liquid metal coolant. If blanket gas is used in the tube, there must be provision to insure its presence in adequate volume, regardless of pressure variations, entrainment, or dissolution in the coolant.

Materials of the tubes must be corrosion-resistant and must maintain their elastic properties at high temperature.

Another type of pressure detector being investigated involves a thin diaphragm exposed to the coolant on one side and maintained in null position by a gas pressure on the other side. The balancing gas pressure may be measured remotely and the null position maintained by a needle valve actuated by the diaphragm so as to control the gas supply. This detector is relatively accurate and simple but the thin diaphragm is a particularly vulnerable part of the coolant system. The diaphragm must be protected from overstress due to pressure surges, and there must be provision for stopping the escape of coolant in case of diaphragm failure.

Use of bellows in pressure detecting devices is avoided wherever possible because of their susceptibility to failure from corrosion and fatigue.

Although there are pressure limits which must not be exceeded for safe operation, the response time of pressure detectors need not be unusually short. Normal operation of the plant does not involve rapid fluctuations in pressure.

3. Temperature. Temperatures may be measured by means of thermocouples and resistance thermometers. Those measuring coolant temperatures are mounted in wells which must be tight, strong and corrosion-resistant. Thermal contact with the ambient coolant must be good so that heat generated by absorption of neutrons and gamma rays in the sensitive element will be dissipated to maintain the element at the temperature of the coolant to be measured.

Electrical insulation of thermocouple wires in the radiation zone must be such as to resist damage from neutron and gamma radiation and high temperature. There is evidence that many organic insulations deteriorate rapidly in such environments. It is believed that quartz, glass or ceramic insulations will give reasonably good service.

Measurements of temperature and pressure in the reactor core and fuel elements are important in determining the allowable power rating and observing the condition of the reactor in much the same way that the temperatures and pressures in the combustion chambers of an internal combustion engine are related to its power rating and condition. Clever means, such as temperature-sensitive paints and piezoelectric pressure detectors have been devised for internal combustion engines, and similarly clever means of detection eventually must be worked out for the reactor.

4. Flow. Operation of the experimental reactor will involve a wide range of coolant flow rates. This makes it difficult to measure flow with fixed orifice flow meters.

Flow of liquid metal coolant may be measured over a wide range by electromagnetic flow meters.* This type of flow meter has the very great advantage that it does not require penetration of the pipe wall or introduction of fixed or moving devices into the coolant stream.

The electromagnetic flow meter involves a permanent magnet to produce magnetic flux through the liquid metal flowing in the pipe. Motion of the conducting liquid metal in the magnetic field causes an electric potential gradient

* Alexander Kolin, *Rev. Sci. Instr.* 16, 109 (1945).

normal to the magnetic field and direction of flow. This potential is measured by means of electrodes attached to the pipe. For a given magnetic field, the potential measured is proportional to the flow of liquid, provided the flow is only a function of radius in the pipe.

There is considerable practical difficulty in calibrating any type of liquid metal flow meter for the large flows and high temperatures encountered in the power plant, so for some time to come the calibration of these meters may be by extrapolation from calibrations of small meters. However, the device has good sensitivity and stability when changes in magnetic field and electrical conductivity due to temperature are taken into account. It is believed that the magnetic field will not be affected appreciably by irradiation of the magnet.

5. Liquid level. It is necessary to detect the level of the liquid metal coolant in storage tanks, surge tanks and in a number of other locations. Level indicators which involve transmitting motion or force from a float through the wall of the vessel are of questionable reliability if they involve immersed moving parts, thin diaphragms or bellows.

Electrical contacts through which the liquid metal closes an electrical circuit upon attaining a certain level may be used in some locations. These have the possible difficulty that condensing metal vapor may, in time, short-circuit insulation of the contact. Other means, utilizing electrical properties of the liquid metal, may be developed.

6. Leak detection. It is vitally important that leaks in the liquid metal system be detected and located at their very incipience. Means for doing this usually involve detecting the metal vapor. The mercury vapor detector* used in coal-fired mercury boilers is an example of this type of detection. In the atomic power plant the problem is considerably more difficult in the radioactive zone. It is not possible to locate the leak by inspection, once it has been detected, so there must be compartmentation of the space around the liquid metal system and numerous leak detectors to localize the trouble.

7. Optical systems. There must be optical systems for viewing locations that are inaccessible because of radiation. Conventional glass mirrors and lenses are blackened in time by exposure to radiation, and plastic lenses cannot withstand the high temperatures, so optical systems must employ polished metal reflecting surfaces where exposed to radiation, and heat in and around the reactor.

Conclusion

As may be inferred from this very meager and incomplete discussion, much remains to be done in the development of instruments for operating and maintaining atomic power plants of the type described here.

Perhaps the biggest obstacle to development is the lack of instrument testing facilities. There are only a few places in the country where the effect of radiation on materials and devices may be studied. Similarly, there are only a few places where liquid metal systems are available for instrument testing. Nowhere is there a combination of radiation and high temperature liquid metal such as will exist in this power plant. More and better testing facilities are being developed, however, so work on instrumentation will be accelerated.

Operation of the first plant may be limited somewhat by inadequacy of instrumentation, but this first operating experience will direct attention to the most critical problems and probably will reveal limitations beyond those discussed here.

* T. T. Woodson, *Ind. Med.* **10**, 22 (April, 1941).

Safety Criteria in Radioactive Water Monitoring*

With the development of the nuclear reactor and with the increased use of radioactive materials in research laboratories, this subject takes on increased importance. In this paper, safe values for the maximum permissible counting rates from radioactivity contaminated water samples, are established under various conditions. Criteria are obtained which are valid for unknown mixtures of contaminating isotopes, based on an additivity rule for the hazards from the separate isotopes. Consideration is given to the use of information limiting the amount of the most dangerous isotope(s) present, and to the modification of the monitoring procedure by means of absorbers or by radioactive decay. The various criteria developed are illustrated by numerical examples based on an arbitrarily calculated set of permissible doses.

METHODS OF TESTING WATER for radioactive contamination are likely to be needed on an increasing scale in the future. This paper considers the basic principles involved. The problem is to develop monitoring procedures which are valid despite the limitation of a paucity of preliminary information on what radioactive isotopes are present and in what proportions.

Additivity of Hazards

A considerable amount of attention has been given to fixing maximum permissible levels for the activity of individual isotopes in, for example, air or drinking water. In water monitoring, however, we are generally concerned with mixtures of isotopes of varying and uncertain composition. Fortunately there is a simple rule applicable to mixtures which, although often unnecessarily stringent, is almost certainly safe.

Let σ_i denote the specific activity of an isotope i, and let t_i denote the maximum permissible value of σ_i. Define $h_i = \sigma_i/t_i$ as the hazard due to i. Then the rule is simply that *the sum of the hazards from all the isotopes present must not exceed unity.*

$$\sum \frac{\sigma_i}{t_i} = \sum h_i \leq 1 \qquad (1)$$

This can be established as follows: Ideally, the presence of i at its maximum permissible level t_i implies that some animal or plant tissue will receive its maximum permissible radiation dose rate (often taken as 0.5 rem/week), and that other tissues will receive less. If, therefore, i is present at a level σ_i, the radiation dose rate in any one tissue may be expressed as a fraction $\alpha_i h_i$ of the maximum permissible, where α_i is unity in the tissue most affected, and is less than unity elsewhere. For a mixture of isotopes, the total dose rate in any one tissue is then a fraction $\Sigma\alpha_i h_i$ of the maximum permissible. Hence we require

$$\Sigma\alpha_i h_i \leq 1 \qquad (2)$$

in each and every tissue.

To employ the whole set of inequalities (2), one for each tissue which might be affected, would be difficult. Since, however, the α_i's never exceed unity,

$$\Sigma\alpha_i h_i \leq \Sigma h_i$$

from which it follows that (1) is a sufficient safety criterion.

* The authors wish to thank the director of the Atomic Energy Research Establishment for permission to publish this paper.

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Equation* 1 is of very wide application. It is immaterial, for example, whether some of the i's are α-emitters and others are β-emitters; or whether some of the t_i's are for drinking water, others are for immersion, and others again are for fish. All that is assumed in the deduction is that the physiological effects of two or more isotopes on any tissue which they irradiate are additive.

Obviously, too, the validity of (1) is unimpaired if, for one or more of the σ_i's, we can only set an upper limit, or for one or more of the t_i's, a lower limit.

Although we shall employ (1) throughout this paper, its unnecessary stringency in comparison with (2) should be noted. Its use implies, for example, that if one isotope is present in maximum permissible quantity, then all others must be completely excluded. Yet the body could quite well tolerate, say, radioiodine (which concentrates in the thyroid) and radiocobalt (in the liver and kidneys), both at very nearly unit hazard. The α's for these elements are, in fact, small in all but the particular organs of the body in which they concentrate.

Dilution Factors

It often happens that the water under test is diluted with large amounts of uncontaminated water before it has a chance to cause any harm. Discharge into a river, for example, involves a considerable dilution factor. If the dilution factor is d, the effect is to raise the upper limit for the total hazard from unity to d, so that (1) must be replaced by

$$\Sigma h_i \leq d \qquad (3)$$

Assessment of Hazards from Practical Tests

If an instrument could be devised which responded to different isotopes in inverse proportion to their t_i's, then criterion (1) or (3) could be applied directly to water monitoring. The nearest approach might be a living animal. For practical purposes, however, no such instrument exists.

Criterion (1) or (3) could also be applied directly if all the separate σ_i's were determined by radiochemical analysis. This would be impracticable as a routine procedure, though one or two particularly dangerous isotopes might be isolated and measured individually.

Chief reliance is therefore placed on tests which can be used to establish upper limits for the hazards from groups of isotopes. α-emitters can be assessed as a group, for example, and so can hard β-emitters. Provided the tests cover all isotopes which may make an appreciable contribution to the total hazard, then it is possible to apply the additivity rule (1) or (3). Indeed it is often convenient to allocate definite fractions g_I, g_{II}, g_{III}, etc., of the total hazard to the different groups, so that $\Sigma_I h_i \leq g_I d$, $\Sigma_{II} h_i \leq g_{II} d$, $\Sigma_{III} h_i \leq g_{III} d$ $\cdots$ etc., where $g_I + g_{II} + g_{III} \cdots \leq 1$.

Consider a test in which a response n_i—usually a counting rate—is obtained from an isotope i at specific activity σ_i. The efficiency f_i for i may be defined by the equation

$$n_i = f_i \sigma_i \qquad (4)$$

and will be a function of the geometry,

* "Equation" is used as a generic term. Actually, many of the relations so noted in this paper are "inequalities."

and of the energy of the radiations emitted by i, etc. Similarly, for a mixture of isotopes

$$n = \sum n_i = \sum f_i \sigma_i \qquad (5)$$

Some of the f_i's will usually be zero, or at least very small, *e.g.*, a test for α-emitters will give no response with β-emitters; but they will never be negative.

Now let N_i be the counting rate resulting from i present at activity t_i:

$$N_i = f_i t_i$$

(so that $h_i = n_i/N_i$), and let the isotopes be arranged in order of their N_i's:

$$N_1 \geq N_2 \geq N_3 \geq \cdots N_r \cdots \geq N_s \qquad (6)$$

Then

$$\sum_1^r h_i = \sum_1^r \frac{n_i}{N_i} \leq \sum_1^r \frac{n_i}{N_r} \leq \frac{n}{N_r} \qquad (7)$$

This inequality shows that n/N_r represents an upper limit for the hazard due to the group of isotopes 1, 2, 3 . . . r. If r were the only isotope present the hazard would in fact be equal to n/N_r.

The safety criterion corresponding to (7) is

$$n \leq gdN_r \qquad (8)$$

where g is the fraction of the total hazard allocated to the group 1, 2, 3, . . . r. Equation 8 combined with (7) implies $\sum_1^r h_i \leq gd$, as required.

The argument given is perfectly general, provided all the f_i's are positive, and shows that in *any* group it is the isotope with the smallest N_i that sets an upper limit to the hazard, and may therefore be regarded as the most dangerous of the group. It follows that (6) arranges the isotopes in order of increasing danger. Note particularly that the N_i's, which are counting rates, rather than the t_i's themselves, are the significant quantities; for monitoring purposes high counting efficiency may offset high toxicity.

Use of Information on the Most Dangerous Isotope (s)

Should (7) prove too stringent, *i.e.*, should it yield too high an upper limit for the hazard, then further information must be sought if the water under test is not to be rejected forthwith as unsafe. Such information must relate directly or indirectly to the most dangerous isotope or isotopes. Three different cases may be considered:

1. σ_r is known, *e.g.*, from a radiochemical analysis for r. In this case, a simple modification of the argument leading to (7) yields:

$$\sum_1^r h_i \leq \frac{n - f_r\sigma_r}{N_{r-1}} + \frac{\sigma_r}{t_r} \qquad (9)$$

which is superior to (7) except when r

LIST OF SYMBOLS

B	background count
d	dilution factor
D	standard deviation of count
f_i	counting efficiency for isotope i
g	fraction of total hazard allocated to particular group of isotopes
h_i	hazard due to isotope i
k_i	factor limiting proportion of activity due to isotope i
n	total count from sample
n_i	count due to isotope i
N_i	count due to isotope i when present at its maximum permissible concentration
${}^1N_i$, ${}^2N_i$, *etc.*	See Equation 14, *etc.*
t_i	maximum permissible concentration of isotope i
T	counting time or time of decay
λ_i	disintegration constant of isotope i
σ_i	specific activity of isotope i in sample

is the only isotope present. The corresponding safety criterion is

$$n \leq gdN_{r-1} - f_i\sigma_r\left(\frac{N_{r-1}}{N_r} - 1\right) \quad (10)$$

If the next most dangerous isotope $(r-1)$ has also been determined, then (10) may be extended to:

$$n \leq gdN_{r-2} - f_{r-1}\sigma_{r-1}\left(\frac{N_{r-2}}{N_{r-1}} - 1\right) - f_r\sigma_r\left(\frac{N_{r-2}}{N_r} - 1\right) \quad (11)$$

and similarly for an estimation of the three, four . . . *etc.*, most dangerous isotopes.

2. σ_r does not exceed a known upper limit, σ_r'. Such a limit might, for example, be fixed from a knowledge of the maximum turnover rate of r in an establishment. In this case it can be shown that (9) and (10) are still valid with σ_r' in place of σ_r, since the coefficient of the σ_r term on the right-hand side of (10) is negative.

3. σ_r does not exceed a known fraction of the total specific activity, *i.e.*, $\sigma_r \leq k_r \sum_1^r \sigma_i$. It should usually be possible to fix a limit of this kind for r, especially if r is a relatively rare isotope.

First, let $\sigma_r = k_r \sum_1^r \sigma_i$. Then it may readily be shown that

$$\sum_1^r h_i = \sum_1^{r-1} \sigma_i[1/t_i + \{k_r/(1-k_r)\}1/t_r]$$

$$n \geq \sum_1^r f_i\sigma_i = \sum_1^{r-1} \sigma_i[f_i + \{k_r/(1-k_r)\}f_r]$$

These equations are of the same form as those from which (7) was derived, *viz.*,

$$\sum_1^r h_i = \sum_1^r \sigma_i/t_i \quad \text{and} \quad n \geq \sum_1^r f_i\sigma_i$$

Hence, by analogy

$$\sum_1^r h_i \leq n/{}^1N_q \quad (12)$$

and

$$n \leq gd\,{}^1N_q \quad (13)$$

where

$${}^1N_i = \frac{f_i + [k_r/(1-k_r)]f_r}{1/t_i + [k_r/(1-k_r)]1/t_r} \quad (14)$$

and q is the isotope with the smallest ${}^1N_i$ (other than r itself). Now let $\sigma_r < k_r \sum_1^r \sigma_i$; then it may be shown that (12) holds *a fortiori*.*

Equation 12 can be extended as follows: Let $\sigma_q \leq k_q \sum_1^r \sigma_i$ in addition to $\sigma_r \leq k_r \sum_1^r \sigma_i$

then $\sum_1^r h_i \leq n/{}^2N_p$ and $n \leq gd\,{}^2N_p$

where $${}^2N_i = \frac{f_i + \dfrac{k_q f_q + k_r f_r}{1-k_q-k_r}}{\dfrac{1}{t_i} + \dfrac{k_q/t_q + k_r/t_r}{1-k_q-k_r}}$$

and p is the isotope with the smallest ${}^2N_i$ (other than q and r). Now p may be limited in its turn by a relation $\sigma_p \leq k_p \sum_1^r \sigma_i$ giving a ${}^3N_p$, and so on. The proof in the general case is somewhat complex, and will not be given here.

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* Let $\sigma_r = k_r' \sum_1^r \sigma_i$, where $k_r' < k_r$, and let ${}^1N_i'$ be the value of ${}^1N_i$ when k_r' replaces k_r. Then it may readily be shown that ${}^1N_i' > {}^1N_i$. But by the argument just given $\sum_1^r h_i \leq n/{}^1N'_{q'}$ where q' is the isotope with the least ${}^1N_i'$. Then

$$\sum_1^r h_i \leq n/{}^1N'_{q'} \leq n/{}^1N_q \leq n/{}^1N_q$$

(If q' is identical with q, there is one stage fewer in the argument.) Equations 12 and 13 are therefore valid for $\sigma_r \leq k_r \sum_1^r \sigma_i$.

It is important to note that (12) and its extensions are still valid when some of the f_i's vanish, *i.e.*, when some isotopes are not counted at all in the tests used. Provided upper limits can be fixed for the proportions of these isotopes it is still possible to fix a safe maximum counting rate.

Modifications to the Test

Additional information may also be obtained by modifying the test. There are in fact three simple modifications readily carried out in practice, *viz.*,

1. Insertion of an absorber between the test specimen and the counting instrument;
2. Reduction of the thickness of the specimen;
3. Leaving the specimen to decay radioactively.

The first two modifications have an important feature in common: for every isotope,

$$f_i \geq f_i' \qquad (15)$$

where the prime refers to the modified test. Equation 15 is usually valid in case (3) also, though there are occasional exceptions when growth of a radioactive daughter occurs.

Inequalities of the form of (7) and (8) will apply to the modified test. Since, however, the order of the f_i'/t_i products may be different from that of the f_i/t_i products, some other isotope r', instead of r, may determine the maximum permissible counting rate:

$$\sum_1^r h_i \leq n'/N'_{r'} \text{ and } n' \leq gdN'_{r'} \qquad (16)$$

This criterion will be superior to Equation 8 provided

$$n'/n < N'_{r'}/N_r$$

Whenever (15) is valid, a further inequality can be deduced. We have

$$n - n' = \Sigma(f_i - f_i')\sigma_i$$

in which all the terms on the right are positive. Hence,

$$\sum_1^r h_i \leq (n - n')/(N_{r''} - N'_{r''}), \quad \text{and}$$

$$n - n' \leq gd(N_{r''} - N'_{r''}) \qquad (17)$$

where r'' is now the isotope with the least value of $N_i - N_i'$ in the group 1, 2, 3, . . . r. Equation 17 will be superior to (8) when

$$n'/n > 1 - (N_{r''} - N'_{r''})/N_r$$

Consider, for example, modification (3). In the absence of growth of daughter isotopes,

$$f_i' = f_i \exp(-\lambda_i T)$$

where λ_i is the disintegration constant of i, and T is the time. Equations 16 and 17 become, respectively,

$$\sum_1^r h_i \leq n'/N_{r'} \exp(-\lambda_{r'} T), \quad \text{and}$$

$$n' \leq gdN_{r'} \exp(-\lambda_{r'} T)$$

$$\sum_1^r h_i \leq (n - n')/N_{r''}[1 - \exp(-\lambda_{r''} T)],$$

and

$$n - n' \leq gdN_{r''}[1 - \exp(-\lambda_{r''} T)]$$

If, as is nearly always the case, r is a relatively long-lived isotope, the former of these criteria will usually be superior to (8), while if r is short-lived the latter might be preferable.

Similarly, if the radiations from r are relatively hard, it may pay to insert an external absorber and use criterion (16), and if they are soft, to take the difference between counts with and without an absorber, and use criterion (17).

Against all such modifications must be set, however, a loss in sensitivity, resulting from the smaller counting rates in the modified tests.

Sensitivity Limits

Monitoring procedures frequently have to be carried out at low activity levels, where statistical fluctuations in counting set a limit to the sensitivity.

If $n + B$ is the observed counting rate of the test sample, B the observed background count, and T the time for which each is measured, then the standard deviation D of the net counting rate n is given by

$$D = \sqrt{(2B + n)/T}$$

For small values of n, this is approximately equal to $\sqrt{2B/T}$.

Now the sensitivity limit is reached when D amounts to some fraction of n. The selection of a suitable value for this fraction is arbitrary, but 0.2 is a reasonable choice. The least value of n to be regarded as significant is then $5\sqrt{2B/T}$. This value of n (or n') may be inserted in any of the safety criteria in previous sections to show whether the test will yield significant results. When (8) is used, for example, the requirement is

$$(gdN_r/5)\sqrt{T/2B} \geq 1 \qquad (18)$$

The quantity on the left-hand side of (18) forms a convenient measure of the sensitivity of the test.

When criterion (17) is used, it is not necessary to take a background count. The least significant value of $n - n'$ is $5\sqrt{\frac{n + n'}{T}}$ corresponding to a sensitivity of $[gd(N_{r''} - N'_{r'})/5]\sqrt{T/(n + n')}$ (In this expression only, n and n' are the actual counting rates without subtraction of the background.)

APPLICATIONS

Permissible Dose

Although the formulas for calculating the t_i's for different isotopes in, for example, drinking water are well-known, there is still much uncertainty as to the numerical values of many of the biological factors. The t_i values may be in doubt by one or more orders of magnitude.

Many of the refinements in the present paper might therefore seem unnecessary. However, exact compliance with legally fixed t_i values may be enforced. If so, it is important to establish the least stringent safety criterion applicable in any particular case. A saving of a factor of two or three may mean a saving of thousands or even millions of gallons in the amount of water which has to be handled.

TABLE 1

Calculated Values of t_i and N_i

Isotope	t_i ($\mu\mu c/l$)	f_i (*cpm per* $\mu\mu c/l$)	N_i (*cpm*)
[C^{14}	0.24×10^{3}	5.2×10^{-3}	1.2]
[Na^{22}	0.29	97	28]
P^{32}	20	240	4800
S^{35}	43	6.7	290
Ca^{45}	13	20	260
Sc^{46}	5.1	34	170
Fe^{59}	13.5	42	570
Co^{56}	2.3	220	510
Co^{60}	0.21	27	5.7
Sr^{89}	6.5	220	1400
$Sr^{90} \rightarrow Y^{90}$	0.024	94	2.2
In^{114}	4.3	270	1200
Sb^{124}	3.8	180	680
I^{131}	2.8	84	230

As no generally agreed list of t_i values exists, a series has been calculated (see Table 1) on more or less arbitrary assumptions, purely in order to illustrate the use of the safety criteria developed in this paper. The assumptions are as follows:

1. There is no excretion of activity by the body.
2. The activity is uniformly distributed over a 1,000 gm sphere of tissue (radius taken as 6.2 cm). An exception is made in the case of I^{131}, for which the mass of the thyroid (taken as 25 gm) is used.
3. The daily water intake is 4 l.
4. The maximum permissible dose is 0.5 rep/week over a lifetime of 70 years.

The nuclear data required have been

taken from Marinelli, Quimby and Hine.*

It is to be emphasised that the f_i figures in Table 1 have no validity as "true" permissible-dose values. Nevertheless it is believed that the assumptions made are on the whole pessimistic, and that conclusions based on them will be safe, often by a large margin.

The list of isotopes in Table 1 is identical with that given by Marinelli, Quimby and Hine, but omits *K*-capture isotopes (whose counting efficiencies are not yet known very certainly) and isotopes with periods of less than a week. C^{14} and Na^{22} have been placed in brackets and left out of further consideration because they would in reality be almost completely excreted.

Monitoring Procedures

Routine monitoring procedures must be simple and rapid, employing reasonably robust instruments, yet at the same time sufficiently sensitive. The following methods have been developed at the Atomic Energy Research Establishment.

α-activity of the total solids. In this test, a 25 ml water sample is evaporated to dryness leaving a thin layer of solid residue, 0.75 mg/cm² thick, on a platinum tray 10 cm² in area. The tray is inserted under a scintillation α-counter which has a background count of approximately 0.2/min.

β-activity of the total solids. Up to 1 l of water is evaporated to dryness under alkaline conditions, and the solids are spread on a tray 3 cm² in area, giving a layer up to 100 mg/cm² thick. Under normal conditions the only β-emitters likely to be lost are isotopes of the inert gases which are of little danger in drinking water. The tray is inserted under a cylindrical end-window Geiger-Müller counter which has a window thickness of 7 mg/cm² and a background count of 10/min. Under these conditions the efficiency of β-ray counting falls off with decreasing β-ray energy, but only the very softest β-rays are missed altogether. The efficiency for γ-rays is low. The f_i values in Table 1 are for this test, assuming the full 100 mg/cm² thickness.†

The immersion counter. As a guide to the β- and γ-activity levels of water in the various parts of an effluent system, a cylindrical immersion Geiger-Müller counter is very convenient. It is sensitive to hard β-rays and to γ-rays. It can give direct readings without any chemical manipulation, and is suitable for use with automatic recording devices. Unfortunately the walls cannot be made thin enough to pass soft β-rays, so the test misses such isotopes as C^{14} and S^{35} for which the f_i's are zero. There is a further disadvantage in the rather high background count (80/min), which limits the sensitivity.

Chief reliance for monitoring purposes is placed on the tests which measure the α- and the β-activity of the total solids. For simplicity, the total permissible hazard is divided equally between the α- and the β-emitters, *i.e.*, the g-factors for the two tests are put equal to 0.5. If an appreciable hazard were to arise in the future from other groups of isotopes, *e.g.*, pure γ-emitters, it might be necessary to introduce further tests with appropriate g-factors.

Use of the Safety Criteria

The use of the safety criteria developed in the early part of this paper will be illustrated principally with reference to the second of the tests just mentioned, the measurement of the β-activity of the total solids.

* L. D. Marinelli, Edith H. Quimby, G. J. Hine, NUCLEONICS **2**, No. 4, 56 (1948).

† The thickness varies a little from day to day, owing to variations in the solid content of the water. It would be possible, but laborious, to calculate the true f_i's for all thicknesses. It is simpler, and safe, to reduce the f_i's for the greatest thickness used (100 mg/cm²) proportionally to the actual thickness.

The simple total solids test. The fourth column of Table 1 gives the N_i values for this test, obtained by multiplying the t_i's by the f_i's. The lowest N_i is that for Sr^{90}, which is therefore the most dangerous of the isotopes listed. Criterion (8), with $g = 0.5$, then gives $n \leq 1.1d$.

The sensitivity of the test, from (18), is $0.049d\sqrt{T}$. For the direct monitoring of drinking water, for which $d = 1$, T must be greater than 412 min, if the sensitivity is to exceed unity. Clearly therefore, with the t_i figures in Table 1, this simple unmodified test is not quite sensitive enough for drinking water.

For effluent which is to be diluted by a large factor, the test is amply sensitive. For $d = 100$, for example, a few minutes counting suffices. With so much sensitivity in hand, it may be expedient to reduce the volume (1 l) of the initial water sample. A reduction to 0.1 l, for instance, decreases the maximum permissible counting rate and the sensitivity by a factor not exceeding 10.

Radiochemical analysis. The simple test just described is a very stringent one. To employ criterion (8) is tantamount to assuming that the entire contamination is due to Sr^{90}, the most dangerous isotope. If $\sigma_{Sr^{90}}$ is determined (in $\mu\mu c/l$) by radiochemical analysis, then the less stringent criterion (10) may be employed, with Co^{60} setting the limit:

$$n \leq 2.9d - 0.15\sigma_{Sr^{90}}$$

If in turn $\sigma_{Co^{60}}$ is determined, a much greater advantage accrues, because the next N_i value, that for Sc^{46}, is higher by an order of magnitude: $n \leq 85d - 0.78\sigma_{Co^{60}} - 7.2\sigma_{Sr^{90}}$. The sensitivity is thereby sufficiently increased to enable the test to be used for drinking water.

Use of limits for the most dangerous isotope(s). Radiochemical analysis for Sr^{90} can be avoided if it is known that not more than, say, 1% of the contamination consists of this isotope. Criterion (13) can then be employed, with $k_r = 0.01$.

TABLE 2

Calculated Values of ^1N_i and ^2N_i

Isotope	N_i (*cpm*)	^1N_i ($k_{Sr^{90}} = 0.01$)	^2N_i ($k_{Sr^{90}} = k_{Co^{60}} = 0.01$)
P^{32}	4800	510	460
S^{35}	290	17	16
Ca^{45}	260	42	38
Sc^{46}	170	57	52
Fe^{59}	570	87	79
Co^{56}	510	260	240
Co^{60}	5.7	5.2	—
Sr^{89}	1400	390	350
$Sr^{90} \rightarrow Y^{90}$	2.2	—	—
In^{114}	1200	420	390
Sb^{124}	680	270	250
I^{131}	230	110	100

The ^1N_i values calculated on this basis are given in Table 2. The least is that for Co^{60} and the maximum safe counting rate is $2.6d$. This compares with $1.1d$ for the simple test. The gain in sensitivity is just sufficient to make the test useful for drinking water; when $d = 1$, $T = 80$ min gives a sensitivity of unity. There is again a much greater advantage when Co^{60} is brought into consideration. If this isotope, like Sr^{90}, is limited to 1% of the total, we obtain the set of ^2N_i's given in column three of Table 2. The least ^2N_i is that for S^{35} (not, it may be noted, Sc^{46}, which comes next after Sr^{90} and Co^{60} in the order of N_i's), and the maximum safe counting rate is $8.0d$ for drinking water. The test is now amply sensitive for drinking water.

Modifications to the test. Table 3 shows the effects of (*a*) inserting 27 mg/cm^2 of aluminum between the test specimen and the counter, and (*b*) leaving the test specimen to decay for 2.6, 30 and 42 days.

In the external absorber test it is obvious that criterion (16) is of no

TABLE 3

Calculation for Modified Tests

Isotope	*Unmodified Test*	*External Absorber Test (27 mg/cm²)*		*N_i'—Decay Test*		
	N_i	N_i'	$N_i - N_i'$	*2.6 days*	*30 days*	*42 days*
P^{32}	4800	3800	1000	4200	1200	650
S^{35}	290	0	290	280	230	210
Ca^{45}	260	0	260	260	230	220
Sc^{46}	170	0	170	170	130	120
Fe^{59}	570	0	570	550	370	310
Co^{56}	510	390	120	500	400	360
Co^{60}	5.7	0	5.7	5.7	5.7	5.6
Sr^{89}	1400	1100	300	1400	960	830
$Sr^{90} \rightarrow Y^{90}$	2.2	1.1	1.1	5.8	9.4	9.4
In^{114}	1200	950	250	1200	790	710
Sb^{124}	680	530	150	660	480	420
I^{131}	230	93	140	180	17	5.9

value for the elements listed, because some of the N_i''s vanish. As regards criterion (17), Sr^{90} has the smallest $N_i - N_i'$ and is therefore still the most dangerous isotope. The criterion is $n - n' \leq 0.55d$, which may hold even if the simple criterion $n \leq 1.1d$ fails.

In the decay test there is a complication due to growth of Y^{90} from Sr^{90}. After 2.6 days, growth of Y^{90} is sufficient to bring N_i for the $Sr^{90} \rightarrow Y^{90}$ pair up to the value for Co^{60}, which thereafter becomes the most dangerous isotope. The safety criterion (16) is then $n' \leq 2.8d$, involving an increase in sensitivity by a factor of 2.6 over the unmodified test, which is sufficient to enable the new test to be applied to drinking water. Moreover, if there are short-lived isotopes in the test sample, the counting rate after 2.6 days, n', may be appreciably less than the initial rate, n. The safety criterion may therefore be less stringent than the criterion in the unmodified test.

Up to 42 days, the safety criterion remains $n' \leq 2.8d$ and any decrease in n' which occurs during that time will improve the test. Thereafter, however, the relatively short-lived I^{131} sets the limit, and nothing will normally be gained by further periods of decay.

In view of the possible growth of Y^{90}, criterion (17) is not valid for the decay test unless applied to decay occurring in the later stages after growth of Y^{90} is complete. Even so, however, the values of $N_i - N_i'$ for such long-lived isotopes as Co^{60} are very small, rendering the criterion of little or no value.

CONCLUSIONS

The numerical discussion in this paper has been given solely in order to illustrate the principles involved. The detailed conclusions are valid only for the particular list of isotopes considered, and for the arbitrary permissible-dose values used. For this reason it does not yet seem profitable to investigate all the various combinations of tests and safety criteria that are possible.

In practice the circumstances of any particular establishment will determine which isotopes must be considered and which are the most suitable monitoring

procedures to use. Where the contamination is due to β-emitters and is of fairly constant radioactive composition, *e.g.*, with atomic power plant effluent, the immersion counter may prove an adequate monitor. Soft β-emitters which are missed by such counters can be dealt with by means of Equation 13 and its extensions. In other cases the total solids tests are recommended. Individual isotopes which are counted inefficiently may be dealt with either by radiochemical analysis, or again by means of Equation 13 and its extensions.

It should be noted that it is not necessary to know the tolerances for all contaminating isotopes before the methods of this paper can be applied, but only those for the most dangerous. A cursory examination of a table of nuclear data will generally show whether a particular isotope is likely to require consideration, and effort can then be concentrated on obtaining biological data for the few of interest.

Finally, attention is drawn to the great simplicity for the routine certification of water of the criteria developed here. All take the form of maximum permissible counting rates. Although it is a matter of some complexity to arrive at sound values for these counting rates, once they have been obtained the operator is only called on to certify whether or not they have been exceeded.

NO ONE CAN SPELL OUT the future in detail. but in the field of atomic energy alone, we have possibilities for peaceful development that are almost beyond imagination. I rather think that atomic energy today is about where the electrical industry was at the time of Faraday, and that the development of its use will have at least as great an impact on society. We know full well that material progress for two hundred years has always been associated with the increase in available capital and available power, permitting the substitution of machines for human effort. First, the steam engine, then hydroelectric engines and gas engines, made progress possible. Now we have this new and almost unbelievably great source of energy rapidly becoming available for use. The military implications of atomic energy have thrown a temporary cloud of secrecy over this development, but already the use of radioisotopes, as a research tool, gives promise of great things, making possible new discoveries in all fields of knowledge that mean a major advance in science and technology. The international situation may delay progress temporarily, but the day will surely come when the peaceful use and application of this new, and fantastically large, source of energy from atoms, will certainly open up a new and potentially great field of engineering, technology, industry, and opportunity.

—From an address given by C. D. Howe, Trade Minister of Canada, before the engineering graduates of McGill University (May 27, 1949)

Interim Recommendations:

Disposal of Radioactive Wastes by Non-AEC Users*

ALL BIOLOGICAL SYSTEMS are constantly irradiated with penetrating ionizing radiations. These radiations originate from (1) cosmic sources, (2) naturally occurring radioactive materials, and (3) cosmic-ray-induced radioactivity.

As a result of natural and cosmic-ray-induced radioactivity, most all substances, including the human body, contain minute quantities of radioactive materials. The quantities of radioactivity present in the atmosphere, in the ocean, in the soil and rocks, and in biological media vary considerably with location and other factors.

Exclusive of the radioactivity present in a biological system, the system is always subject to both cosmic rays and to the gamma rays from the radioactive materials naturally present in the environmental media. The combination of these two produces a natural background radiation which, at sea level, is usually greater than or equal to 0.2 mr per 24 hours. Background radiation increases with altitude because of an increase in cosmic-ray intensity. Added to the varied environmental or background radiation will be the radiation intensity in the biological material due to various amounts of radioactivity which are ever-present in biological systems (for example, small quantities of radium, radon, uranium, thorium, thoron, potassium-40, and carbon-14). The control of human body irradiation is thus a matter of degree. Exposure or possible exposure of the body to radiation must hence always be discussed in quantitative terms.

The total radiation from background radiation and naturally present internal radioactivity may be considered as the radiation "floor," and the maximum permissible exposure value, that is, the maximum continued exposure rate at which significant deleterious effects are not known to be produced, may be taken as the radiation "ceiling." In selecting methods of disposal of radioactive material, exposures that may result from such disposal should be kept as far below the "ceiling" as is possible within the realms of practicability.

BASIC CONSIDERATIONS

After examining the physical characteristics of an isotope and its chemical and biological behavior, quantitative criteria can be set up for disposal by either of two general methods:

A. By dilution and dispersion

1. *Dilution with stable isotopes of the same element in the same chemical form*

   When the activity per gram of element, or specific activity, is reduced by dilution with stable isotopes of the same element, further concentration of the radioactivity is obviously impossible without isotope separation. To insure uniform dilution, the stable isotopes used as the diluent should not only be of the same element, but should also be in the same chemical form. Since biological systems have no mechanism for appreciable separation of isotopes, further concentration in nature will not take place. In fact, the specific activity is most likely to be further reduced by dilution with stable isotopes in the environment.

2. *Dilution with media other than stable isotopes of the same element*

   The radioactivity per unit volume of matter may also be reduced to

* This material was issued as Circular B-6 by the Isotopes Division, Atomic Energy Commission, P. O. Box E, Oak Ridge, Tenn.

safe proportions through dilution with various media, such as air and water. The maximum permissible concentration for each isotope is determined for the diluting medium used. These values are calculated on the basis of activity in the media *per se*. In contrast with diluting a radioisotope with stable isotopes of the same element, the possibility must be considered of reconcentration of the activity in surrounding media by chemical or physical means or by biological systems. This is particularly true when water is used as the diluent. A special case of dilution is discussed below under "Confinement and Control."

B By confinement and control

1. *Dilution and confinement*

   One special case of a dilution medium is the use of concrete, the active material being contained in the liquid used to mix the concrete. The hardened concrete is finally segregated by storage or burial on land or at sea. Burial may be justified on the assumption that the material will be eroded away only after a considerable lapse of time and then at a slow rate; furthermore, such activity as may still exist in the eroded material will be subject to further dispersion.

2. *Concentration and confinement*

   a. Some methods of concentration:

      To facilitate the handling of disposable radioactive materials, it is often desirable to reduce the bulk through a convenient method of concentration. Some methods which have been employed to concentrate active materials in liquids are evaporation, precipitation, and ion exchange. Radioactivity contained in materials other than liquids, particularly biological materials, may often be concentrated by incineration. The latter sometimes affords a means of concentrating both volatile and non-volatile materials. In the process of incineration non-volatile materials are left in the ashes and volatile materials may be removed from the effluvium through scrubbing. The volatile materials may be allowed to escape without scrubbing provided dispersion keeps the concentration below the maximum permissible value. In all cases particulate material of high specific activity should be removed from the effluvium.

   b. Some methods of confinement:

      Concentrated materials of short half-life may be stored until the activity decays to a value which will allow the material to be disposed of by dilution and dispersion, or until it has decayed to a negligible value. If materials of long half-life are disposed of by confinement and segregation, they can be diluted with stable isotopes of the same element in the same or equivalent chemical form to the extent that safe conditions are realized if and when the segregated material is released from confinement. The case of confinement by concrete has already been noted. When radioisotopes are buried in the ground, one must consider the possibility of transfer through leaching, ion exchange, or other chemical or physical mechanisms. Consequently, care must be exercised in the selection and maintenance of the burial ground.

NEED FOR INTERIM RECOMMENDATIONS

The disposal of radioactive wastes and radiocontaminated materials is a problem to some recipients of radioisotopes outside the Atomic Energy Commission laboratories. The problem has been accentuated due to lack of formal authoritative recommendations in this field. This deficiency has been recognized by the National Committee on Radiation Protection. Recently this committee established a Subcommittee on Waste Disposal and Decontamination. This subcommittee is to study the broad problems of disposal and make recommendations which are applicable to the disposal by off-Commission users of all radioactive materials, natural, cyclotron-produced and pile-produced. Authoritative sources estimate that one and one-half years will be required for the subcommittee

to prepare these recommendations.

It was felt that although disposal of radioactive wastes from laboratories not controlled by the Atomic Energy Commission presents no serious problem at present, some recommendations should be formulated to serve during the interim period in which the above subcommittee is studying the broad phases of disposal problems. Consequently, the AEC Isotopes Division, in cooperation with the Division of Biology and Medicine of the AEC Washington office, called a meeting in Washington September 20, 1948, for consideration of this problem. Leading authorities (see Appendix) in attendance at this meeting formulated interim recommendations for disposal of the radioisotopes which are likely to be used in greatest quantity in off-Commission facilities during the interim period. Procedures recommended were based upon the simplest methods of disposal that will insure protection of personnel and prevention of contamination. These recommendations were formulated with the understanding that they will be modified and supplemented as dictated by program expansion and disposal experience, and that they will eventually be superseded by recommendations of the Subcommittee of the National Committee on Radiation Protection.

Because of the immediate need for specific methods of disposal of the most widely used radioisotopes, the interim recommendations were confined primarily to methods of disposal of iodine-131, phosphorus-32 and carbon-14, which together account for 90% of the activity of radioisotopes shipped to date.

INTERIM RECOMMENDATIONS

I. Disposal of Radioiodine

A. Recommended procedures:

1. Radioiodine may be discharged from an institution into the main sewer, provided that:
   a. The daily volume of water flowing from the sewage outlet of the institution to the main sewer is sufficient to dilute the radioiodine to 0.5 microcurie per liter;
   b. Maximum activity disposed of from any one institution will not exceed 200 millicuries per week;
   c. Regular radiation surveys are made of plumbing fixtures;
   d. Appropriate surveys are made before repairing the plumbing between the disposal outlet and the main sewer;

or

2. Radioiodine, as excreted from patients or in the form of iodide, may be discharged from an institution into the main sewer, provided that:
   a. To each millicurie of radioiodine discharged, one gram of potassium iodide is added at the time of disposal;
   b. The daily average volume of water flowing from the sewage outlet of an institution to the main sewer is sufficient to dilute the radioiodine to 10 microcuries per liter;
   c. Regular radiation surveys are made of plumbing fixtures;
   d. Appropriate surveys are made before repairing the plumbing between the disposal outlet and the main sewer.

B. Some considerations in the formulation of procedures:

1. For ingestion the maximum permissible concentration in water is calculated to be 24 microcuries per liter.
2. It is impractical to store excreta contaminated with radioiodine in large institutions because of the excessive volume, malodor, and unnecessary exposure to laboratory personnel.
3. A concentration of 0.5 microcuries per liter was considered safe for body surface exposure for any individuals who might come in contact with the effluent.
4. The upper limit of 200 millicuries per week is considered safe, and this value is sufficient for disposal from most institutions using radioiodine at present.
5. Iodine is excreted primarily as

iodide; consequently, dilution of the radioactive excreta with an iodide such as potassium iodide should insure proper chemical form for homogeneous mixing.

6. With the specific activity given by the addition of one gram of potassium iodide per millicurie of iodine, the maximum concentration in the thyroid possible by drinking the sewage water would be approximately 0.1 microcurie per day. This is based on the normal thyroid uptake of 100 micrograms of iodine per day. Assuming constant uptake at this rate, the equilibrium or maximum concentration in the gland would be 1.15 microcuries.

II. Disposal of Phosphorus-32

A. Recommended procedures:

1. Radiophosphorus may be discharged from an institution into the main sewer, provided that:
   a. The daily volume of water flowing from the sewage outlet of the institution to the main sewer is sufficient to dilute the radiophosphorus to 0.1 microcurie per liter;
   b. Each millicurie is diluted with 10 grams of phosphorus as phosphate at the time of discharge. (Note: The stable phosphate considered in the dilution may be both material naturally and artificially supplied);
   c. The maximum activity disposed of in any one institution does not exceed 200 millicuries per week;
   d. Appropriate radiation surveys are made before repairing the plumbing between the disposal outlets and the main sewer;
   e. The sewage does not enter directly into fresh water systems.

B. Some considerations in the formulation of procedures:

1. The maximum permissible concentration in water for ingestion and for body submersion is 4.8 microcuries per liter and 1.4 microcuries per liter, respectively.
2. Because P^{32} in fresh water may be concentrated 10^5 fold by some microorganisms, it was considered pertinent to lower the permissible concentration to 0.1 microcurie per liter. Dilution by 10 grams of phosphorus as phosphate insures a form suitable for homogeneous mixing and reduces the specific activity to a level which should prevent pronounced reconcentration by any organism.
3. The effluent should not be discharged directly into fresh water because the phosphate content of the fresh water may be too low to prevent significant reconcentration by biological systems. Conversely, the phosphate content in the usual sewage system should insure a reduction in specific activity through high dilution with the stable element.

III. Disposal of Carbon-14

A. Recommended procedures:

1. Radiocarbon may be exhausted in the air, provided that:
   a. No person shall be exposed to the inhalation of air containing greater than 10^{-8} microcuries per cc;
   b. Particulate matter is filtered from the exhaust air.

B. Some considerations in the formulation of procedures:

1. A concentration of 10^{-8} microcuries per cc of air is considered safe for inhalation.
2. Due to the possible implications of overexposure to the lungs from minute particles of high activity, it was felt that particulate matter containing C^{14} should not be discharged into the air.

IV. Disposal of Any Radioisotope

As explained above, it will require study over a period of years to derive disposal procedures to use for each radioisotope under all the various circumstances that may arise. Certain possible procedures were brought up for discussion as follows:

A. Dilution with stable element to a very low rate of energy dissipation

1. Radioisotopes of any half-life may be buried in the earth if they are uniformly diluted with stable isotopes of the same chemical element and in the same chemical form, to the extent that 4.15 ergs are dissipated per gram of element per

day,* provided that:

a. The burial is made only in suitably selected areas which are in possession of, and will be maintained by, the user;

b. The material is buried at a minimum depth of 5 feet.

2. Some considerations in the formulation of procedures:

a. Materials at the above elemental dilution could not, under any circumstances, be concentrated to greater than maximum permissible values in biological material.

b. Burial at a depth of 5 feet should be both practical and safe provided care is exercised in selecting the site for the burial ground.

B. **Dilution with stable element to a specific activity below a value which would not permit the body to reconcentrate the activity above permissible values.** To illustrate:

1. Suppose that the permissible level of P^{32} for the organ of maximum concentration (skeleton) is 17 microcuries and that this organ contains 530 gm of phosphorus. Then, assuming uniform distribution, each gram of stable phosphorus would contain 0.032 microcuries of P^{32}, or a ratio of 31.3 gm of stable isotope per microcurie of radioisotope. Thus, phosphorus containing P^{32} in this ratio, even if it were continually taken into the body, could not produce greater than maximum permissible exposure to any part of the body.

2. Suppose that the permissible value of I^{131} for the organ of maximum concentration (thyroid gland) is 0.87 microcuries and that this organ contains 0.02 gm of iodine. Then, assuming uniform distribution, each gram of stable iodine would contain 44 microcuries of I^{131}, or a ratio of 0.023 gm of stable isotope per microcurie of radioisotope. Thus, iodine containing I^{131} in this ratio, even if it replaced all the iodine in the organ, could not produce greater than maximum permissible exposure.

3. Suppose that the permissible value of C^{14} for the whole body is 983 microcuries and that the body contains 12,950 gm of carbon. Then, assuming uniform distribution, each gram of stable carbon would contain 0.0759 microcuries of C^{14}, or a ratio of 13.2 gm of stable isotope per microcurie of radioisotope. Thus, carbon containing C^{14} in this ratio, even if it were continually taken into the body, could not produce greater than maximum permissible exposure to any part of the body.

C. **Possibility of dilution in concrete and segregation** (See "Basic Considerations," p. 22)

The interim group was not able to give full consideration to the establishment of quantitative criteria for these possible procedures in the case of all radioisotopes involved. However, it was agreed that procedure IV.A.1 above could certainly be considered on the safe side.

These and other procedures will be considered by the Subcommittee on Disposal and Decontamination of the National Committee on Radiation Protection. It is anticipated that the recommendations for methods of radioactive waste disposal from this committee will be much more extensive than these interim recommendations.

The Interim Committee in session in Washington on September 20, specifically passed recommended procedures given as Items I, II, III, and IV.A. The discussion presented on these recommendations, as well as other background material included in this paper, was prepared by the staff of the Isotopes Division.

APPENDIX

Participants in Meeting on Waste Disposal of Radioactive Materials

L. F. Nims, Director, Division of Biology, Brookhaven National Laboratory
K. Z. Morgan, Director, Health Physics Division, Oak Ridge National Laboratory
H. M. Parker, Director, Health Physics Division, General Electric Company, Hanford
F. P. Cowan, Director, Health Physics Division, Brookhaven National Laboratory
J. E. Rose, Director, Division of Health Physics, Argonne National Laboratory

* This rate of energy expenditure is equivalent to 50 mrep in the medium per 24-hr period.

William F. Bale, Professor of Radiation Biology, University of Rochester
Hymer L. Friedell, Director, Department of Radiology, University Hospitals, Western Reserve University
Edith Quimby, Department of Radiology, College of Physicians and Surgeons, Columbia
Robley D. Evans, Department of Physics, Massachusetts Institute of Technology
A. H. Holland, Jr., Director, Office of Research and Medicine, Oak Ridge Operations
Bernard Wolfe, Medical Advisor, New York Operations
Carl Braestrup, Physics Laboratory, City of New York Department of Hospitals, and Consultant, Radiological Laboratory, New York Operations
Paul C. Aebersold, Chief, Isotopes Division, Oak Ridge Operations
Lauriston S. Taylor, Chief, Health Physics Branch, Division of Biology and Medicine, Washington
Spofford English, Chief, Chemistry Branch, Division of Research, Washington
John Z. Bowers, Asst. Director, Division of Biology and Medicine, Washington
Oliver H. Townsend, Office of Public Information, Washington
A. E. Gorman, Sanitary Engineer, Division of Engineering, Washington
Commander E. W. Hirbar, U. S. N., Office of Military Application, Washington
Major Maxwell Dauer, Office of Military Application, Washington
J. H. Jensen, Division of Biology and Medicine, Washington
Harry G. E. Stoeckle, Jr., Acting Chief, Biology and Medicine Division, Office of Research and Medicine, Oak Ridge Operations
N. H. Woodruff, Asst. Chief, Isotopes Division, Oak Ridge Operations
G. W. Morgan, Asst. Chief, Advisory Field Service Branch, Isotopes Division, Oak Ridge Operations

National Committee on Radiation Protection

THE FOLLOWING INFORMATION is taken from the preface to the booklet, "Medical X-ray Protection up to Two Million Volts," which is Handbook 41 of the National Bureau of Standards (March 30, 1949):

The Advisory Committee on X-ray and Radium Protection was formed under the sponsorship of the National Bureau of Standards and with the cooperation of the leading radiological organizations upon the recommendation of the International Commission for Radiological Protection. This Committee, although small in size, has functioned effectively. However, the advent of atomic energy has introduced a large number of new and serious problems in the field of radiation protection.

At a meeting of the Advisory Committee in December, 1946, the representatives of the various participating organizations agreed that the problems in radiation protection had become so manifold that the Committee should enlarge its scope and membership, and should appropriately change its title to be more inclusive. Accordingly, at that time, the name of the Committee was changed to the National Committee on Radiation Protection. At the same time the number of participating organizations was increased and the total membership considerably enlarged. In order to distribute the work load, seven working subcommittees were established as noted below. Each of these committees is charged with the responsibility of preparing protection recommendations in its particular field. The reports of the subcommittees are approved by the main committee before promulgation.

The following individuals representing the organizations indicated, compose the main committee:

H. L. Andrews and E. G. Williams, U. S. Public Health Service; S. L. Warren, Shields Warren, and K. Z. Morgan, AEC; L. F. Curtiss and L. S. Taylor, National Bureau of Standards; E. E. Charlton and L. L. Call, National Electrical Manufacturers Association; H. B. Williams, and A. U. Desjardins, American Medical Association; R. S. Stone, and G. Failla, Radiological Society of North America; R. R. Newell, and J. L. Weatherwax, American Roentgen Ray Society; and L. S. Taylor, International Commission for Radiological Protection.

The following are the subcommittees, with the name of the chairmen:

(1) Permissible External Dose, G. Failla; (2) Permissible Internal Dose, K. Z. Morgan; (3) X-rays up to Two Million Volts, H. O. Wyckoff; (4) Heavy Particles (Neutrons, Protons, and Heavier), Dean Cowie; (5) Electrons, Gamma Rays, and X-rays Above Two Million Volts, L. Marinelli; (6) The Handling and Disposal of Radioactive Isotopes and Fission Products, H. M. Parker; and (7) Monitoring Methods and Instruments, H. L. Andrews, acting chairman.

ABSOLUTE BETA COUNTING*

Although many persons have studied the comparison method of absolute beta counting using end-window Geiger tubes, no thorough studies appear in the literature describing the procedures and results in detail. Having made such a study, the author has made the results available to others who might wish to do absolute beta counting.

This paper deals with the following topics:

***1.* The detailed procedure for determining the corrected efficiency (often called geometry) of an end-window Geiger counter using radium E obtained from discarded radon tubes.**

***2.* The determination of the effects of absorption and scattering on the counting rates of Co^{60} (0.3 Mev), for I^{131} (0.6 Mev), for RaE (1.17 Mev), and for P^{32} (1.72 Mev).**

***3.* A study of the various factors that affect backscattering and a listing of the backscattering corrections for many common sample mountings.**

***4.* The quantitative effects of lateral and vertical displacements of the source on beta counting rates.**

***5.* The preparation of RaE sources of known disintegration rate.**

***6.* The detailed procedure for the determination of the absolute disintegration rate of beta emitters using a RaE standard.**

MOST BETA COUNTING with Geiger-Müller tubes involves the measurement of relative counting rates. However, absolute disintegration rates are needed for dosage determinations in medical work, isotope production, fission yield studies, half-life determinations of long lived isotopes and other studies in the physical sciences.

The counting rate of a beta emitter relative to the disintegration rate depends upon the geometry (the percent of solid angle about the source subtended by the sensitive volume of the counter), and absorption of the radiations by the air and window of the counter and the scattering from the source support and assembly. Zumwalt (*1*) has shown that, if the proper corrections are made for absorption and scattering, the geometry of an end-window GM tube is independent of the maximum beta energy between 0.3 and 2.3 Mev. In his work the sources were mounted on 1 mil polystyrene films and the scattering corrections were of the order of 1%. An expression for the counting rate may be written as the product of the corrected efficiency (*i.e.*, geometry), the disintegration rate and a number of factors expressing the effects of resolving time losses, absorption and scattering. The following equation is similar to the one used by Zumwalt.

$$\text{cpm} = (\text{dpm}) \cdot f_W \cdot f_C \cdot f_A \cdot f_B \cdot f_H \cdot f_S \cdot E \quad (1)$$

where cpm = observed counts per minute.

dpm = absolute number of disintegrations per minute.

f_C = factor for the effect of the finite resolving time of the GM tube and circuit.

* Research work done at Brookhaven National Laboratory under the auspices of the Atomic Energy Commission.

By BENJAMIN P. BURTT*
Brookhaven National Laboratory, Upton, New York

f_W = factor for the effect of the beta-particle absorption by the counter tube window and air in the space between the tube and source.

f_A = factor for the effect of air in scattering beta particles into the counter (separated for convenience from effect due to air absorption).

f_B = factor for the increase in counting rate due to backscattering by the material supporting the source.

f_H = factor for the effect of the source support structure and walls of the housing in scattering beta particles into the counter tube.

f_S = factor for the effect of the mass of the source in causing both scattering and absorption of beta particles.

E = corrected efficiency (*i.e.*, the ratio of the counting rate to the disintegration rate for a hypothetical weightless source in which there is no backscattering and no absorption between the source and the sensitive volume of the counter).

To determine the disintegration rate of a beta emitter each of the terms in Eq. 1 must be evaluated. Some are unity in certain counting arrangements; others vary with the beta energy. For maximum beta energies above 0.6 Mev the backscattering factor, f_B, is practically constant for a given material as measured in different counters.

PRELIMINARY EXPERIMENTS

Losses due to the finite resolving time of the GM tube and circuit. The resolving time is the shortest interval in which two successive particles can produce separate discharges in a GM tube. Significant errors are introduced into measurements involving counting rates of over several thousand per minute. While most counting rates in this work were 5,000 cpm or less and the correction small, an approximate determination of the observed counting loss was made by the standard procedure of the multiple addition of small sources. Four sources were counted individually and together, yielding a correction of $0.5 \pm 0.2\%$ per 1,000 counts per min. For the counter used in this work

$$f_C = 1/[1 + (\text{cpm})\ (5 \times 10^{-4})]$$

Backscattering of RaE beta radiation ($E_{max} = 1.17$ Mev). Geiger-Müller counter tubes with end windows of thin mica contained in a lead housing with a conventional Lucite source support were used. See Fig. 1. The internal diameter of the cathode was 2.2 cm. The door of the lead chamber was lined with one-fourth inch Lucite. The samples were thus surrounded on four sides and the bottom by Lucite. Samples mounted on various materials were placed on the removable Lucite shelves.

The sources covering a circular area of approximately 1 cm² were mounted

* On leave of absence from Chemistry Dept., Syracuse University, until September 1.

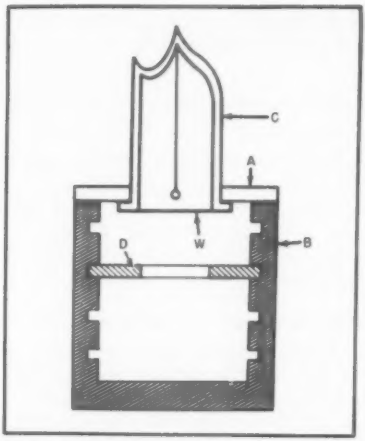

FIG. 1. Counter assembly. *A*) **aluminum top plate;** *B*) **Lucite shelf holder;** *C*) **counter;** *D*) **Lucite shelf containing 1¼ in. hole cut out of center;** *W*) **counter window**

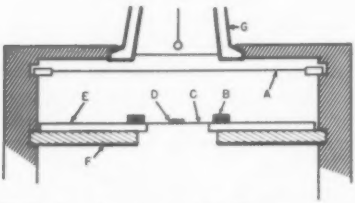

FIG. 2. Counting of RaE sources on thin Zapon films. *A*) **aluminum absorber to remove RaF alpha particles;** *B*) **brass ring supporting Zapon film** *C*; *D*) **source;** *E*) **cardboard;** *F*) **Lucite shelf;** *G*) **counter**

on thin Zapon films (Fig. 2) having negligible backscattering.* The increase in counting rate due to backscattering was measured by observations of the counting rate with and without a solid material behind the film. The film (approximately 20 $\mu g/cm^2$) was mounted on the under side of a brass ring 1¼ inches in diameter and the ring was placed on a piece of 1 mm cardboard with a 1⅜ inch hole cut out of the center. One or two drops of RaE-RaD solution (in radioactive equilibrium) were evaporated on the films and the counting rate determined through a 5 mg/cm^2 aluminum absorber to remove RaF alpha particles. After the activity was determined for this source with negligible backscattering, the cardboard containing the hole was replaced by a solid sheet of cardboard, aluminum or other mounting material and a count again taken. Care was taken to see that the distance from the source to the counter remained the same in all measurements. From these data, after the usual corrections for background and resolving time were made, the backscattering factor f_B was calculated.

$$f_B = \frac{\text{cpm with backing}}{\text{cpm with no backing (Zapon film)}} \qquad (2)$$

The backscattering factors for several mountings are shown in Table 1. The increase in counting rate ranges from 10% for paper to 79% for tungsten and is appreciable even for the lightest of conventional mountings.

The technique was varied slightly for the determination of the backscattering for the 2 mg precipitate on filter paper mounted on cardboard. A standard $Bi(NO_3)_3$ solution containing an aliquot of radioactive bismuth solution (RaE with RaD) was precipitated as $BiPO_4$, filtered in a Hirsch funnel, washed, dried and prepared for counting by placing the filter paper and precipitate on a cardboard mounting. The activity observed when corrected to 100% chemical yield divided by the activity of an aliquot of RaE on a Zapon film is the backscattering factor. Unusual results are noted with nickel. For the 5 mil nickel plate on cardboard $f_B = 1.52$, on aluminum 1.48 and for the 50 mil nickel plate 1.50. Since scattering from aluminum alone is greater than for cardboard alone it is hard to understand why the back-

* See Appendix A for preparation of Zapon films.

scattering is greater from the nickel on cardboard than from nickel on aluminum. This anomaly has not been explained.

Relation between backscattering and distance from source to counter. The backscattering factors for several mountings are plotted in Fig. 3 as a function of distance from the counter. The backscattering factors are constant in the interval from 3 to 20 mm from the counter with the exception of the value for Pd-Ag at 3.4 mm. There is a slight decrease in the backscattering factor as the distance is increased from 20 to 40 mm. This decrease with distance is not a change in the scattering from the mounting, but is caused by appreciable backscattering from the floor of the counter through the Zapon film as the sample is moved closer to the floor (*i.e.*, the denominator of Eq. 2 increases). This may be shown by the following experiment. The counting rate increases with thickness of backscatterer until the backing is about one fifth of the range of the beta radiation in the substance (*1*, *2*). Here saturation is reached and the counting rate is not changed by further increases in the thickness of the backing. The ratio between the counting rates of two samples mounted on different materials infinitely thick for backscattering was measured as the samples were moved closer to the floor of the shelf holder. The results are shown in Fig. 4. The ratios are constant within the experimental error from 6 to 42 mm from the window (44 to 8 mm from the bottom). The backscattering is independent of distance for the light materials measured. In the case of platinum and the palladium-silver mountings there seems to be an appreciable deviation from

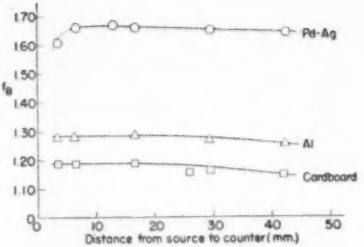

FIG. 3. Backscattering factor as a function of distance from the source to counter for several mountings

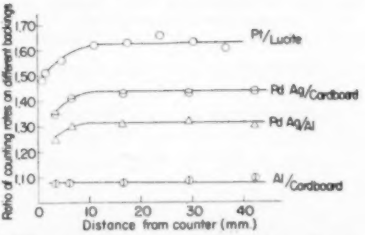

FIG. 4. Ratio of counting rates of two samples mounted on backings infinitely thick for backscattering vs. distance between source and counter window

TABLE 1

RaE Backscattering Factor f_B

(Source on shelf 2, 1.66 cm from the counter window)

Mounting	f_B
Filter paper	1.10
20 mil Lucite	1.15
1 mm cardboard	1.19
2 mg ppt on cardboard	1.19
1 in. watch glass set in 17 mm hole in 1 mm cardboard	1.26
$\frac{1}{32}$ in. aluminum	1.29
5 mil Ni disk on $\frac{1}{32}$ in. Al card	1.48
50 mil Cu	1.48
5 mil Ni plate on cardboard	1.52
50 mil Ni	1.50
50 mil Mo	1.60
Pd-coated Ag disk (used by U. S. Bur. Stds. for RaE standards)	1.66
50 mil Ta	1.75
50 mil Pt	1.78
50 mil W	1.79

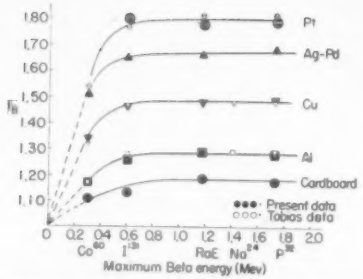

FIG. 5. Saturation backscattering as a function of maximum beta energy for several backing materials

this independence only when the sample is 5 mm or less from the window.

Backscattering of beta radiation of P^{32} ($E_{max} = 1.72$ Mev). The procedure described for RaE was used in obtaining the saturation backscattering factors for P^{32} shown in Table 2. The values of the backscattering factor are independent of distance and the same within the experimental error as those obtained for RaE. Saturation backscattering for P^{32} begins at about 156 mg/cm² and two 1 mm cardboard cards were necessary for saturation. For one card $f_B = 1.14$ and for two or more cards $f_B = 1.18$.

Backscattering of beta radiation of I^{131} ($E_{max} = 0.6$ Mev). This isotope emits gamma rays and this gamma activity must be subtracted from the total observed activity. With the exception of the value on cardboard (see Table 2) the backscattering factors are the same as with beta emitters of greater energy. The factor for cardboard is lower than that observed with P^{32} or RaE.

Backscattering of beta radiation of Co^{60} ($E_{max} = 0.3$ Mev). The backscattering factors for this isotope are shown in Table 2. It is a gamma emitter and the gamma activity was subtracted before making calculations. All of the backscattering factors are lower than those observed with I^{131}, RaE and P^{32}.

Relation of beta backscattering and maximum beta energy. A plot of the saturation backscattering factors for several mountings is shown in Fig. 5 as a function of maximum beta energy. The solid points refer to data obtained in this work and the open points to some results obtained by Tobias (*3*). His results are in good agreement with those obtained by the writer. The internal diameter of the cathode of his tube was 6 mm larger than ours. These data show that the saturation backscattering factor is independent of maximum beta energy above 0.6 Mev. Engelkemeir's data (*2*) show that it is independent from 1.0 to 2.5 Mev.*

Relation of saturation backscattering and atomic number of the backscattering material. A plot of the saturation

TABLE 2

Backscattering Factor f_B for P^{32}, ^{131} and Co^{60} Beta Radiation

Isotope	*Mounting infinitely thick for backscattering*	*Shelf 2*	*Shelf 3*
P^{32}	Cardboard	1.18	1.17
	Al	1.28	1.27
	Ag-Pd	1.68	1.68
	Pt	1.79	—
	Cu	1.49	—
I^{131}	Cardboard	1.14	1.13
	Aluminum	1.26	1.25
	Cu	1.46	1.47
	Ag-Pd	1.65	—
	Pt	1.80	—
Co^{60}	Cardboard	1.11	1.11
	Al	1.17	1.18
	Ag-Pd	1.51	1.53
	Cu	1.34	—

* *Note added in proof:* L. R. Zumwalt reports, in a private communication, that if the backscattering factors are computed at zero total absorber by extrapolation they are approximately independent of energy even below 0.6 Mev.

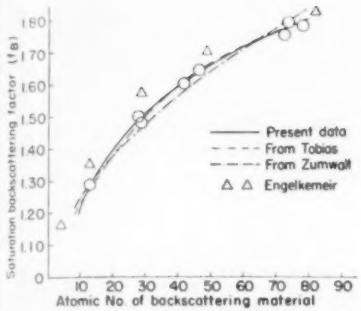

FIG. 6. Saturation backscattering factor as a function of atomic number of backing material

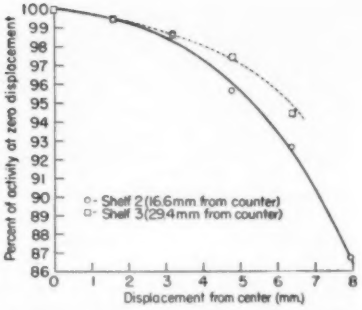
Percent of activity at zero displacement
Displacement from center (mm)
Shelf 2 (16.6mm from counter)
Shelf 3 (29.4mm from counter)

FIG. 7. Counting rate as a function of lateral displacement of sample on shelves 2 and 3

backscattering factors for RaE against atomic number of the backing material is shown in Fig. 6. The circles and heavy solid line indicate data obtained by the writer while the other two curves represent results of Zumwalt (*1*) and of Tobias (*3*) using P^{32}. The triangles are Engelkemeir's (*2*) data. In the latter's work the counter and sample holder were inverted and the backing material placed on top of the source. It is not known whether a shield surrounded his samples or whether they were open to the air on the side opposite the counter. The higher values he obtained could have been due to the difference in the counting arrangement. The results of Zumwalt and of Tobias using aluminum or Lucite-lined lead shields are within 2% of the values obtained by the writer using such a counting arrangement. Engelkemeir's results using the inverted counter are from 1 to 5% higher than the writer's. It is apparent that the values of the backscattering factors for a given material are independent of the end-window counter used within 5%.

Effects on the counting rate of lateral and vertical displacements of the source. To investigate the lateral displacement effect, an aluminum card was cut into strips of different widths and a sample of RaD-RaE solution evaporated on one of the wider strips. The circular sample had a cross-sectional area of approximately 1 cm^2. The activity was measured with the sample centrally mounted. Then the strip containing the activity was displaced laterally and one of the narrow strips moved to fill the gap. This was repeated for other displacements, approximately 100,000 counts being taken for each position. The results for shelves 2 and 3 are shown in Fig. 7 where the counting rate is plotted against the displacement from the central position. On the second shelf (16 mm from the counter) the sample may be displaced as much as 3 mm in one direction without decreasing the observed activity by more than 1.5%. The effect of displacement diminishes as the sample is moved to the lower shelf positions. The sample diameter can therefore be varied by 3 mm without changing the observed count by more than 2%.

While relatively large displacements may be made in the horizontal direction, displacements in the vertical direction cause large changes in the counting rate. On shelf 2, changing the distance from the counter by 1.0 mm will affect the counting rate by 10%. The percentage change in activity per mm

displacement increases to 13% very near the window and decreases slowly to 5%, 50 mm away. A graph of the observed counting rate of a RaE sample against distance from the counter window is shown in Fig. 8. This does not follow the inverse square law until the sample is approximately 70 mm away. Here, according to Blachman's geometry equation,* the deviation from the inverse square law would be of the order of 2%.

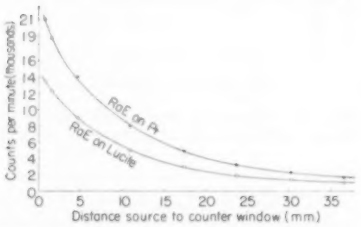

FIG. 8. Observed counts per minute (RaE) as function of distance from source to counter

Effect of the Lucite housing and shelf support structure on the counting rate. It has already been pointed out that for thin mounting materials scattering from the floor of the Lucite shelf support structure makes an increasing contribution to the observed activity as the source is moved closer to the floor. For P^{32} this effect was less than 1% on shelf 2 (16 mm from the counter and 34 mm from the floor) and about 3% on shelf 4 (11 mm from the bottom). Using an aluminum shelf support, Zumwalt (*1*) reported

* See Appendix C.

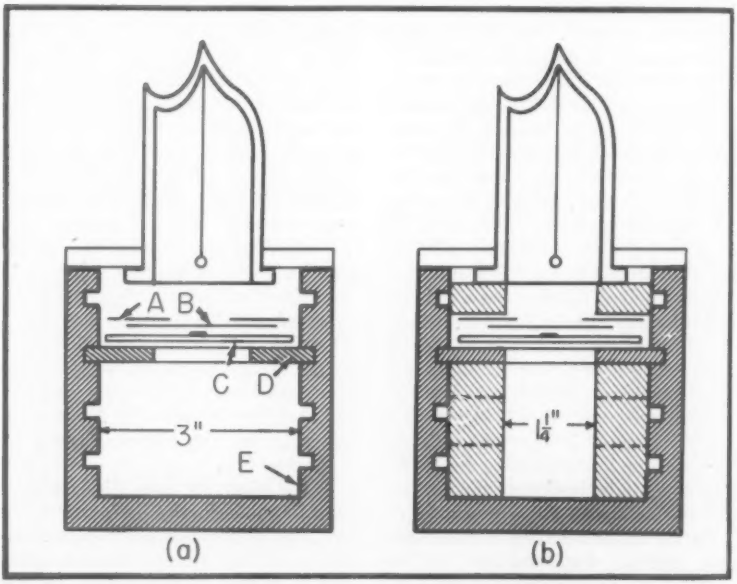

FIG. 9. Arrangement for studying amount of radiation scattered into the counter from the walls. (*a*) **Counter and assembly in first position: *A*, Cu card with 1⅛ in. hole cut out from center; *B*, aluminum absorber; *C*, aluminum card; *D*, Lucite shelf; *E*, Lucite shelfholder.** (*b*) **Assembly with Lucite added**

increases of the order of 0.7% on shelf 2 and 5% on shelf 4.*

A study was made of the amount of radiation scattered into the counter from the walls. The change in counting rate produced by decreasing the distance between the walls from 3 to 1¼ inches was investigated. This was accomplished by completely filling the space between sample and counter, and the space below the sample with Lucite slabs containing a center hole 1¼ inches in diameter. Figure 9*a* shows the counter and assembly in the first position. A copper card 2 mm thick and containing a 1⅛ inch center hole is mounted over the sample. Figure 9*b* shows the assembly with the Lucite and the walls 1¼ in. apart.

Samples mounted on thin Zapon films as well as on aluminum were counted first as in Fig. 9*a* and then with various numbers of the Lucite slabs inserted. The copper plate with the hole in the center was placed over the sample and absorber so that beta particles scattered from the outer edges of the absorber would not reach the counter when the Lucite slabs were absent. When the walls were moved closer to the center there would be no decrease in counting rate because of absorption of these scattered radiations. The copper plate also served to minimize the wall scattering when the walls were 3 in. apart.

There was no detectable difference between the observed activities with the walls in the two positions. Both the aluminum backing infinitely thick for backscattering and the Zapon film with no scattering gave the same results. In this assembly, scattering of radiation into the counter by the walls is negligible.

Correction for absorption. The factor f_W for the absorption by the air and window is evaluated by extrapolation of an aluminum absorption curve to zero absorber. An example is shown in Fig. 10, curve *A*. The absorber is placed as close to the window as possible to minimize scattering into the counter from the absorber. The absorber position is quite important and errors greater than 25% may be produced if the absorber is close to the source or the source is covered. See curves *B* and *C* in Fig. 10. The counting rate is increased by scattering when a thin absorbing material is placed near the source. As pointed out by Willard and Johnston (*4*) the closer an absorber to the sample the larger the fraction of the radiation that is intercepted by the absorber. This results in more total scattering. It is observed that more particles are scattered into the counter than are scattered out. This may be partially explained as follows: First, radiation initially within the solid angle subtended by the counter passes through less absorber and scattering is less probable; second, the solid angle subtended by the counter is small and if a fraction of the radiation is scattered out of this solid angle, only a small fraction of the total radiation must be scattered into the counter to return the counting rate to its original value. The counting rate rises with increasing absorber thickness until the increase in counting rate due to scattering into the counter is just balanced by the radiation that is absorbed and scattered out. Novey and Elliott (*5*) have also reported a maximum in absorption curves taken in this fashion. Because of these scattering effects the absorbers should always be placed as close to the window as possible. The source position is not critical unless a covered sample is used and then the source should be placed near the window leaving only enough room for the insertion of the absorbers.

The first portion of the absorption curve is usually linear when the absorber is placed close to the window.

* If the floor is lead, as it is in some commercial assemblies, or the lead door is unlined, increases of the order of 4–8% may be expected even on the upper shelves.

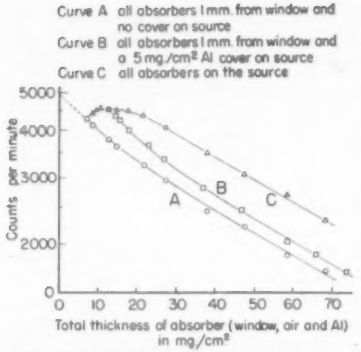
Curve A all absorbers 1 mm. from window and no cover on source
Curve B all absorbers 1 mm. from window and a 5 mg./cm² Al cover on source
Curve C all absorbers on the source
Counts per minute
5000
4000
3000
2000
0
A
B
C
0 10 20 30 40 50 60 70
Total thickness of absorber (window, air and Al) in mg/cm²

FIG. 10. RaE absorption curve extrapolated to zero absorber and the effect of absorber position. Source on Al (1 mm) 1.6 cm from window; thickness of window and air is 7.0 mg/cm².

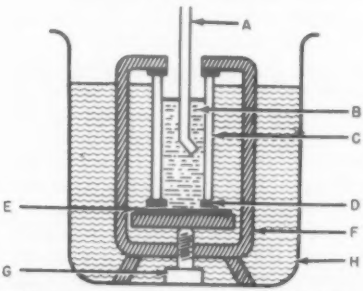

FIG. 11. Assembly for electrolytic deposition of RaE. *A*, mechanical glass stirrer; *B*, RaE solution; *C*, glass chimney; *D*, rubber gasket; *E*, nickel disk; *F*, clamp; *G*, thumb screw; *H*, beaker water bath

The assumption is made that the extrapolated portion of the curve is also linear. The greatest errors in determining disintegration rates probably lie in this step and about 100,000 counts should be taken for each of the initial points on the absorption curve. This may sound extreme but an error of 2% in one of the points may be magnified considerably in the extrapolated value. The absorption factor f_W is the ratio of the counting rate under the experimental conditions to that at zero absorber.

Correction for scattering and absorption by the source. The sources in this work were less than 0.1 mg/cm² and selfscattering and selfabsorption were negligible. When it is necessary to determine the disintegration rate of a source with appreciable weight, the selfscattering and absorption factor f_S must be determined. Sources should be prepared containing the same amount of radioactive material and different amounts of inert carrier. These sources are then counted and the counting rate plotted against the thickness of the source in mg/cm². From this plot the factor f_S can be obtained.

$$f_S = \frac{\text{cpm from thick source}}{\text{cpm from source of zero thickness}} \quad (3)$$

The radiation from sources of finite thickness is both scattered and absorbed. The counting rate may actually increase with sample thickness producing an effect similar to that described for thin absorbers placed on the source. This has been observed in connection with counting the National Bureau of Standards RaD-RaE sources and will be discussed later.

EFFICIENCY OF END-WINDOW COUNTER USING RaE

General Procedure

Radium E is separated chemically from parent RaD and daughter RaF and mounted in such a way that it can be counted in both a methane proportional alpha counter of known efficiency and an end-window beta counter. The beta activity of the RaE and the alpha activity of the RaF as it grows in are plotted against time. From these activities and the known efficiency of the alpha counter the beta disintegration rate can be computed. After making the proper corrections for absorption and backscattering as described above the corrected efficiency is found.

Preparation of RaE Source

1. Separation of RaE from RaD and RaF. Discarded radon tubes were crushed and digested in 0.5*N* HNO_3 for several hours. RaE was extracted from this mixture by the procedure described in detail in Appendix B.

2. Preparation of RaE sample for counting. A 2 ml solution of RaE was placed in a glass chimney clamped on a rubber gasket over a thin nickel plate ($1\frac{5}{16}$ in. diameter and 5 mils thick). It was stirred at 80° C for 75 minutes as shown in Fig. 11. The RaE deposit on nickel is thin, uniform and therefore suitable for accurate alpha counting. The hole in the center of the rubber gasket determines the deposit area.

3. Determination of counter efficiency. The alpha activity of the nickel plate was determined immediately after preparation in a methane proportional alpha counter. The efficiency of this counter for plutonium mounted on platinum is 52.0% and for plutonium mounted on quartz about 50.5% (*6*). These experimental values have been supported by theoretical calculations (*7*). As a check the writer counted plutonium standards mounted on platinum and the efficiency so obtained was 51.9%. It was assumed that the efficiency using the nickel plate was 51%.

The beta activity was determined on shelf 2 through an absorber to remove the alpha particles from RaF. This absorber was placed as near the window as possible. The decay of the RaE and the growth of RaF were plotted against time for approximately three RaE half-lives (15 days). See Fig. 12 for sample data. To correct for slight day to day variations in the counter response a RaD-RaE sample was prepared and counted each day that the decay of the RaE source was measured. This was done by evaporating a drop or two of RaD, RaE solution (in equilibrium) on an aluminum card and covering the activity with a thin Zapon film.

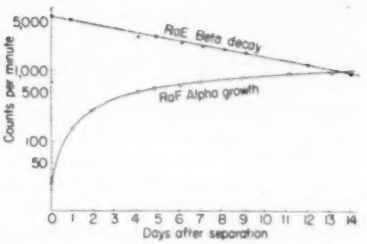

FIG. 12. Decay of RaE and growth of RaF for sample 22

From the known efficiency of the alpha counter and a plot such as that shown in Fig. 12 the beta disintegration rate was computed as follows:

$$D_{1_0} = \frac{A_2}{0.51}\left(\frac{\lambda_2 - \lambda_1}{\lambda_2}\right)\left(\frac{1}{e^{-\lambda_1 t} - e^{-\lambda_2 t}}\right) \quad (4)$$

where t = time after separation of RaE and RaF

D_{1_0} = dpm of RaE at $t = 0$

A_2 = (cpm of RaF at time t) − (cpm RaF at $t = 0$)

λ_1, λ_2 = disintegration constants of RaE and RaF, respectively.

Results

Three samples were used and the results for one of them are plotted in Fig. 12. Letting A_{1_0} equal the extrapolated counts per minute of RaE at $t = 0$ the ratio of A_{1_0}/D_{1_0} was calculated from these curves by means of Eq. 4. These values expressed as percentages are shown in Table 3 for the three samples at two-day intervals after preparation. The values at the end of two days were not used in computing the mean ratio since the alpha activity was taken from an initial steep portion of the curve where there were only one or two experimental points. The mean for $100A_{1_0}/D_{1_0}$ is 9.76. This is the uncorrected efficiency for a RaE source mounted on a nickel plate placed on a 1 mm cardboard. To get the efficiency corrected for absorption and scattering, Eq. 1 must be applied.

For a nickel plate mounted on cardboard and a total of 9.7 mg/cm² of absorber, $f_W = 0.870$. As shown above the factor f_H for the effect of the source support and walls of the housing is equal to 1.00 for shelves 2, 3, and 4 and can be neglected. The factor f_A for the effect of air in scattering beta particles into the counter increases with distance from the tube and with the energy of the beta emitter. For P^{32} at a distance of 5 cm Zumwalt (*1*) reported $f_A = 1.02$. It was assumed in this work using RaE at 1.6 cm that f_A was close to unity. The backscattering factor f_B for a nickel plate mounted on cardboard was 1.52. The sources were essentially weightless and carrier free ($f_S = 1.00$). The corrected efficiency calculated using Eq. 1 is

$$E = \frac{A_1 \times 100}{D_1 \times f_B \times f_W \times f_S \times f_A \times f_H \times f_C} = 7.38\%$$

The RaE standard whose counting rate was known at the time the corrected efficiency was 7.38% can be used to determine the efficiency at other times. The disintegration rate of the standard can be computed from Eq. 1 and used to determine the corrected efficiency of other counters.

To give an idea of the results one can expect with different backings the efficiencies for RaE beta radiation are given in Table 4 in which only the absorption correction has been made. The variations are due to backscattering alone.

DISINTEGRATION RATE OF RaE SOURCES AND OTHER BETA EMITTERS

The disintegration rate of a beta emitter with a maximum energy above 0.6 Mev and mounted on one of the backings listed can be calculated easily from Eq. 1. If the counter has a window diameter of ¾ to 1½ inches and the shelves are mounted in an aluminum or Lucite-lined lead shield the backscattering factor can be obtained from the tables in this article with an uncertainty of 5%. The factor for absorption can be obtained from an extrapolation of an absorption curve to zero absorber, the counting rate is observed and the corrected efficiency obtained by counting a beta standard prepared as described above. Instead of preparing RaE beta standards one can obtain them from the U. S. Bureau of Standards (mounted on palladium

TABLE 3

Ratios of the Observed Beta Counting Rates at $t = 0$ to the Disintegration Rates Computed from Alpha Counting

	$100A_{1_0}/D_{1_0}$		
Days after preparation	*Sample 22* $A_{1_0} = 5950$ cpm $A_{2_0} = 24$ cpm	*Sample 23* $A_{1_0} = 4080$ $A_{2_0} = 10$ cpm	*Sample 24* $A_{1_0} = 5950$ cpm $A_{2_0} = 25$ cpm
2	10.50	10.15	10.81
4	9.67	9.52	9.86
6	9.75	10.10	9.74
8	9.82	9.72	9.70
10	9.82	9.82	9.74
12	9.75	9.82	9.58
14	9.73	9.64	—
Mean of observations (4–14 days) = 9.76			

coated silver) or the efficiency can sometimes be determined by the coincidence method (*8*).

EVALUATION

The question will undoubtedly be raised as to whether the corrected efficiency as defined above is the geometry (the percentage of the radiation that starts out in the direction of the sensitive volume of the counter). It may be, but we prefer to refer to the corrected efficiency only as the ratio of the observed activity to the disintegration rate for a weightless sample mounted without backscatterer and without absorber or window between source and the sensitive volume of the counter. This viewpoint is taken for the following reasons. A calculation of the geometry can be made such as that derived by Nelson Blachman of this laboratory (Appendix C) involving the internal diameter of the cathode, the diameter of the circular sample and the distance between the sensitive volume and the source. The geometry was calculated at different distances from the frame of the counter window and is plotted in Fig. 13. The distance to the frame was used since the exact distance to the sensitive volume was not known. The experimentally determined "geometry" is also plotted in Fig. 13. The curves do not coincide, but if the experimentally determined efficiency is displaced 5 mm to the right the curves match fairly well. This may mean that the sensitive volume begins about 5 mm behind the window frame. The inner edge of the glass bead terminating the anode is about 4 mm from this point. However, agreement could be obtained by assuming that the effective cross-sectional area of the tube is smaller than the physical area. A combination of these two effects would also bring the two curves together reasonably well. It is difficult to measure the distance to the effective volume and

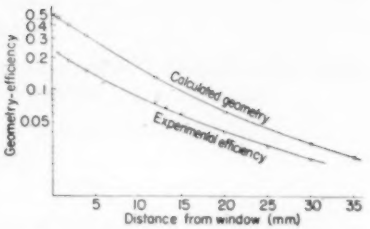

FIG. 13. Calculated geometry vs. distance from counter window; efficiency corrected for absorption and scattering vs. distance from window

a 1 mm error in this measurement makes a 10% error in the corrected efficiency. The experimentally determined efficiency varies slightly from day to day while the true geometry should not vary. The difference probably lies in the ability of the counter to count all those particles which enter its sensitive volume. For the above reasons it is not claimed that the corrected efficiency and the geometry are identical. The usefulness of the corrected efficiency is not impaired by such a viewpoint.

Most of the errors in the determination of the absolute disintegration rate are about 2%. These include such errors as the extrapolation of the beta decay curve to $t = 0$, and the assumed geometry of the alpha counter. There

TABLE 4

Examples of Efficiencies for RaE Beta Radiation Corrected Only for Absorption (measured on shelf 2)

Mounting	Ef_B
Zapon ($\sim 20\mu g/cm^2$)	7.38
Cardboard (1 mm)	8.78
1 in. watch glass (over 17 mm hole in cardboard)	9.30
1/32 in. aluminum	9.52
Cu (50 mil)	10.92
Pd-Ag disk (U. S. Bur. Stds.)	11.81
50 mil Pt	13.14

TABLE 5

Disintegration Rate of RaD-RaE Source No. 10 Determined by Comparison with N.B.S. Co^{60} and RaD-RaE Sources

(Source weight No. 10 < 0.1 mg/cm² mounted on Al card)

Compared with	*Source thickness (mg/cm²)*	*Backing*	*dpm for No. 10*	*Percentage deviation from 62,300 dpm*
1. RaE-RaF calibrated by alpha counting RaF	0.1	Ni	62,300 ± 6%	—
2. RaD-RaE from N.B.S.	1.0 (PbO_2)	Pd-Ag	61,600 ± 6%	1%
3. Co^{60} from N.B.S.	0.1	Zapon	65,500 ± 10%	5%
4. Co^{60} using our coincidence calibration	0.1	Zapon	64,400 ± 10%	3%

may be an error in assuming a linear extrapolation of the RaE absorption curve to zero absorber, and the greatest errors in this method probably lie in the determination of the counting rate at zero absorber. When dealing with beta emitters of energy less than 1 Mev, large extrapolations of the absorption curve are necessary to compute f_W and corresponding errors are introduced. The variations between successive counts are often greater than those predicted from statistics and long counts are necessary. In this work each point was the result of from 50,000 to 100,000 total counts, and the counting error is less than the others.

The disintegration rate of a RaD-RaE source calibrated by the method described above was compared with beta sources calibrated by other methods. Several U. S. Bureau of Standards RaD-RaE sources were used for comparison as well as Co^{60} sources prepared from a solution calibrated by the Bureau of Standards by coincidence measurements. The Co^{60} sources were also checked by coincidence measurements in this laboratory. The results of the comparison are shown in Table 5 after all scattering and absorption corrections have been made. The large uncertainties in the values determined by comparison with the Co^{60} sources are caused by the large extrapolation of the Co^{60} absorption curve to zero absorber.

It should be pointed out that an 8% correction was applied to the counting rates of the N.B.S. RaD-RaE samples that was not applied to the others. This was a correction for selfscattering from the 1 mg/cm² source of PbO_2. The other sources were of negligible thickness and no such correction was needed. That such a correction was needed and that it was of the order of 8% was shown by the following experiments. A RaD-RaE source of negligible weight mounted on a palladium coated silver disk was covered with different thicknesses of lead by vacuum deposition. The counting rate was

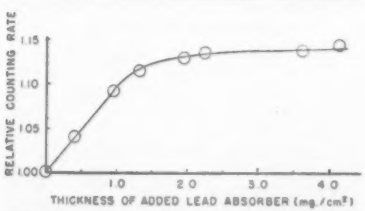

FIG. 14. Lead absorption curve for RaE mounted on a Pd-Ag disk (lead evaporated directly on source)

TABLE 6

Determination of Disintegration Rate of N.B.S. RaD-RaE Sources by Direct Alpha Counting and by Comparison with Author's RaD-RaE Sources

Beta counting rates of N.B.S. sources were divided by a selfscattering factor of 1.08

N.B.S. source no.	*Listed value (dps)*	*By comparison method (dps)*	*By direct alpha counting of old sources (dps)*
82	108	109	105
84	108	109	105
2116	206	206	—
3157	577	582	—
3158	573	581	—

determined after each successive layer had been added. The results of this study are shown in Fig. 14 where the counting rate is plotted against the thickness of lead. This curve was used to compute the increase in counting rate for a RaD-RaE source mounted as 1 mg/cm² of lead on a Pd-Ag disk. It was assumed that the radiation proceeding forward initially passed through 0.5 mg/cm² while the backscattered radiation passed through 1 mg/cm². This computation showed that the counting rate for such a source was 8% higher than a source of negligible weight containing the same amount of radioactive material.

That the correction was of this order of magnitude was also demonstrated by a second experiment. The ratio of the beta counting rate to the alpha counting rate was determined for a N.B.S. RaD-RaE-RaF source that had been prepared three years ago as well as for a "weightless" RaE-RaF source prepared by the author and mounted on the same backing as the N.B.S. source. The ratio for the N.B.S. source was 9% higher than the ratio for our source after making the proper corrections for growth, decay and alpha selfabsorption in the N.B.S. sample. With all other variables held constant the higher beta counting rate for the N.B.S. source must be ascribed to selfscattering. When these selfscattering corrections were made the agreement was very good between the listed values of the N.B.S. sources (corrected for decay) and the values determined by comparison with our sources calibrated independently. The results of this comparison are shown in Table 6 together with the values obtained from a direct alpha count of two old N.B.S. standards after making a correction for the self absorption of the alpha particles.

Whenever the N.B.S. standards are used to determine the disintegration rate of weightless sources a selfscattering correction must be applied unless the source and absorber are placed very close to the window. In the work described above the absorber was in contact with the window while the source was 1.6 cm below the window. The 8% correction is not affected significantly by increasing the distance from the source to counter above 1.6 cm. However, when absorber and source are placed as close as possible to the window, the selfscattering factor becomes unity. These corrections could all be avoided if weightless RaD-RaE sources were issued by the Bureau of Standards and standardized by alpha counting.

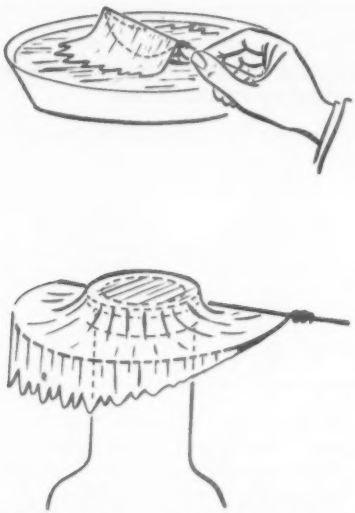

FIG. 15. Preparation of Zapon films

If the disintegration rate of a beta emitter is determined in an aluminum or Lucite-lined lead shield and the backscattering factors listed above are used, the efficiency can be determined within about 6%. If the backscattering factors are determined experimentally somewhat more accurate results may be obtained although others (whom we feel are being over-optimistic) have claimed accuracy of 2% for absolute beta counting.

* * *

The author greatly appreciates the support given to this study by Dr. William Rubinson whose suggestions and comments have been most helpful.

APPENDIX A

Preparation of Thin Zapon Films

Strong films of approximately 10 micrograms per sq cm thickness can easily be prepared from Zapon and thinner. Heavier films can be made by decreasing the amount of thinner used. The films prepared in this work were mounted on a brass ring 1¼ inches in diameter that could be laid over a slightly smaller hole in a cardboard card. The card and ring were placed beneath the window of the Geiger counter.

A solution is prepared by mixing one volume of Zapon with three of thinner. Ten to twenty drops of this solution are placed on the surface of clean water in a dish. The solution spreads to within an inch or two of the edge of the dish and the solvent is allowed to evaporate. A dish at least ten inches in diameter should be used. After two or three minutes, when the solvent has evaporated, a semicircular wire loop is dipped under the water and moved to a position beneath the film. It is lifted, as shown in Fig. 15, draping the film over the wire. In order to transfer the double film to the brass ring, the ring is placed on top of a flask and the wire and film pressed down over the ring. The extra film may be trimmed away with scissors and no gluing is necessary. The film laps over the edge of the ring and when dried remains securely attached. The water may be evaporated from the films in a 110° C oven.

APPENDIX B

Procedure for Separation of RaE from RaD and RaF

Radium E in HNO_3 solution can be separated from RaD by deposition on nickel at 79° C (*9*). Two ml of the RaD, RaE, RaF solution obtained from discarded radon tubes were placed in 45 ml of 0.5*N* HCl, heated to 80° C, and 50 mg of nickel powder (150 mesh) added. After 5 minutes of stirring and centrifugation the nickel was washed with 0.1*N* HCl and the supernate retained for further RaE extraction. The nickel powder was dissolved in aqua regia, centrifuged and any residue discarded. RaE is known to coprecipitate with ferric hydroxide (*10*); 5 mg of ferric ion were added and ammonia

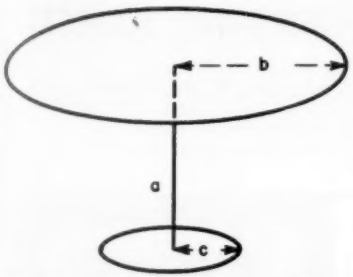

FIG. 16. For calculating geometry. *a*, distance between circles; *b*, radius of larger circle (window); *c*, radius of smaller circle (source)

added in excess. After centrifugation the supernate was discarded and the precipitate washed twice with water, dissolved in HCl, reprecipitated and centrifuged. After the precipitate was dissolved in 8*N* HCl the iron was extracted with three 20 ml portions of isopropyl ether. The remaining water solution contained the RaE in approximately 80% yield together with some RaF.

RaF can be separated from RaE by deposition on silver powder (*11*). Four ml of the RaE, RaF solution were evaporated to 0.05 ml and 2 ml of 0.5*N* HCl added. The RaF was extracted with four 6 mg portions of silver powder. The silver powder mixture was stirred vigorously for three minutes. This was accomplished by placing a small capillary tube in the solution and drawing air through it into the solution with a partial vacuum. The silver was discarded after each extraction and the bubbler washed thoroughly. The resulting solution contained RaE with less than 0.1% of the original RaF. The acid concentration during the silver extraction was kept low to insure good separation. Large amounts of silver were avoided since they also remove appreciable quantities of RaE

APPENDIX C

Calculation of Geometry of Disk Sources and End-Window Counters

The geometry of an end-window Geiger counter is the percent of solid angle about the source subtended by the sensitive volume of the GM tube. The following equation for the geometry was derived by Nelson Blachman of this laboratory. See Fig. 16.

The geometry can be written as a series in c/a:

$$G = 0.5\left[1 - \frac{1}{(1+\beta)^{1/2}} - \frac{3}{8}\frac{\gamma}{(1+\beta)^{5/2}} - \gamma^2\left(-\frac{5}{16}\frac{\beta}{(1+\beta)^{7/2}} + \frac{35}{64}\frac{\beta^2}{(1+\beta)^{9/2}}\right) - \gamma^3\left(\frac{35}{128}\frac{\beta}{(1+\beta)^{9/2}} - \frac{315}{256}\frac{\beta^2}{(1+\beta)^{11/2}} + \frac{1155}{1024}\frac{\beta^3}{(1+\beta)^{13/2}}\right)\right]$$

where $\beta = b^2/a^2$ and $\gamma = c^2/a^2$. This series converges if γ is not too large. β may have any value. Rapid approximations of the geometry with errors of the order of 10% can be made by assuming a point source (*i.e.*, $c = 0$). The geometry equation for a point source is

$$G = 0.50\left[1 - \frac{1}{(1+\beta)^{1/2}}\right]$$

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PINHOLE CAMERA FOR GAMMA-RAY SOURCES*

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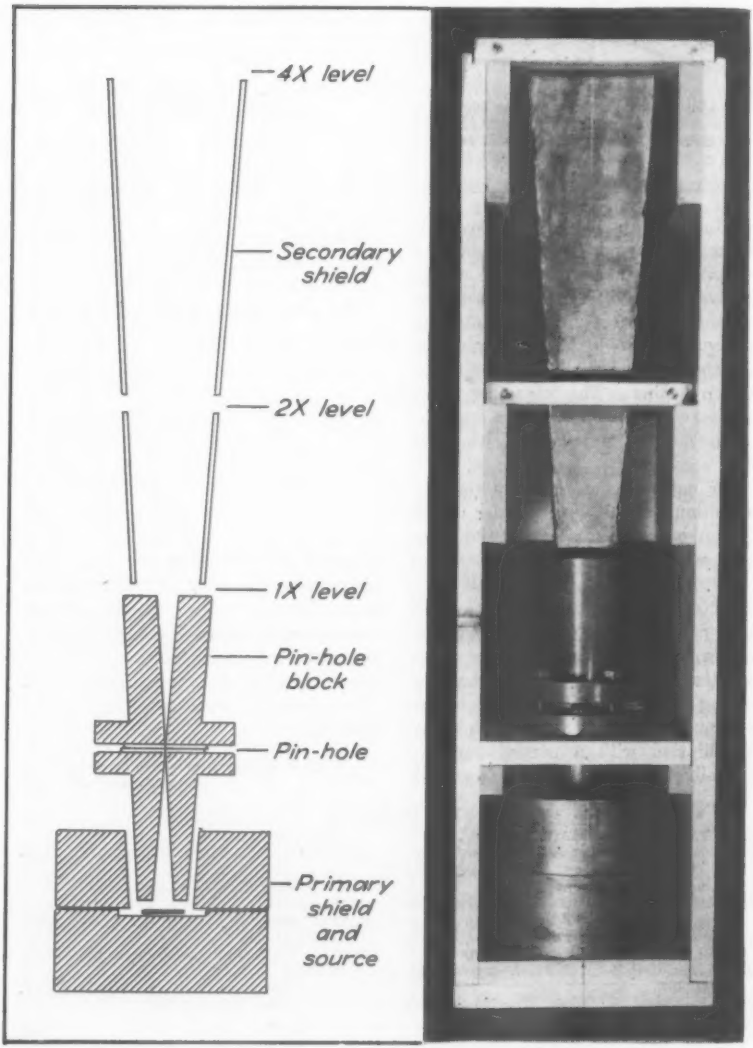


Views a three-quarter-inch field at a distance of five inches from the pinhole. The images obtained indicate the camera's usefulness as a tool where exposure length is not of critical importance and for check on material that cannot be handled as a contact or radioautograph

PHOTOGRAPHY of radioactive sources has been largely confined to radioautographic techniques in which no attempt is made to focus or collimate the rays. The more closely the active source is approached to the recording emulsion, the less the circle of confusion and the more accurate the localization.

Marton and Abelson† introduced a focusing device that is based on the use of a magnetic field, much as it is used in the electron microscope. It is limited by its inherent principles to the focusing of charged particles, notably monokinetic beta rays. Uncharged radiations such as gamma rays are not influenced by the magnetic field.

Since there is at present no known way of bending gamma rays so that they can be focused and photographed in the usual sense, an alternative is the method of selective interference such as is used in a pinhole camera.

Principles of Design

The following may be said about the design of any pinhole camera:

1. The transmitting orifice must be surrounded by opaque, nonpenetrable material.

2. The smaller the orifice, the better is the resolution, and the longer the necessary exposure. In shorter wavelengths the problem of diffraction about the orifice is of less concern.

3. The farther the orifice is from the source, the better the resolution. The farther it is from the recording emulsion, the greater the magnification. In either step the exposure time is increased.

The chief problem in the design of a gamma-ray pinhole camera is the fulfillment of the first principle. Materials are only relatively opaque to gamma activity, their opacity increasing with atomic number and density. Of the commonly available materials, lead has both these desirable characteristics to a high degree.

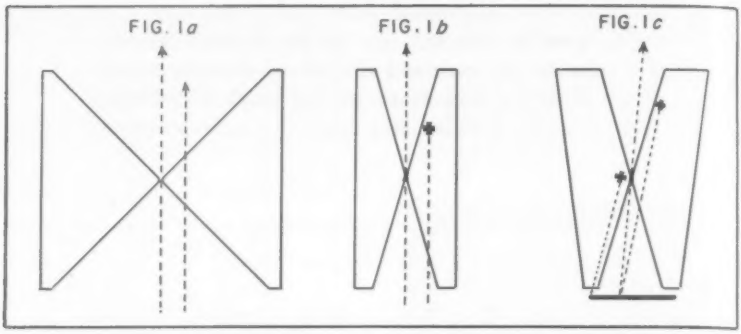
Since lead is only relatively opaque, the next step is to determine the thickness of lead necessary to stop enough of the rays so that the image formed by way of the orifice will have a minimum of background, gamma-induced fog. Assuming a rather high energy of 1.8 Mev, it would take 1.28 cm of lead to stop 50% of the rays‡. It is obvious that such a thickness of lead about a

To remove or insert material, the pinhole camera is disassembled in the following order: *Remove the secondary shields, lift the pinhole block from its seat, and slide the top primary shield off to expose the depression in the lower primary shield*

* A demonstration of the camera was given last year by the authors at meetings of the American Association of Anatomists. The work was supported in part by the H. C. Bumpus Fund.

† L. Marton, P. H. Abelson, *Science* **106**, 69 (1947).

‡ Based on an absorption coefficient of 0.54. W. Heitler, "Quantum Theory of Radiation," fig. 21, p. 216 (Oxford University Press, 1947).



Pinhole camera basic principles. (*a* and *b*)—Comparison of the two figures shows that the narrower the incident and exit cones of the pinhole, the more effectively is the lead arranged to stop any specific strength of gamma ray. (*c*)—Two cones are necessary. The lead about the lower cone stops rays that are not in the photographic field of the camera. That about the upper cone stops rays whose source is in the field but which do not pass through pinhole

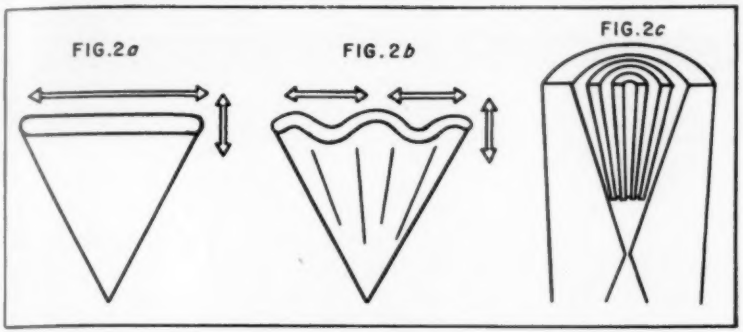
pinhole would not stop enough of the total rays to allow a clear image to be formed. As a minimum thickness, 8.5 cm (approximately 5 inches) was selected. If used in complete theoretical effectiveness, that thickness would give an image (orifice transmission) of 100% to 1% of background fog (shield transmission).

The actual effectiveness of the pinhole and consequent resolution of the camera for any particular gamma-ray strength is directly dependent on the slope of the incident and exit cones. The narrower the cones, the more effectively is the lead arranged. Narrow cones (Fig. 1*b*) provide a much better armoring of the pinhole than do broad cones (Fig. 1*a*). In practice, the narrowing of the cones to gain a more effective pinhole (and consequent enhanced resolution) is limited by the size of the field that the investigator wishes to photograph. A field of ¾-inch diameter was chosen for the camera illustrated in order that it could view centrally placed material on an ordinary micro-slide.

It is necessary that the incident and exit cones be identical and that they each extend the predetermined "stopping thickness" of lead from the pinhole, in this case 5 inches. The 5 inches of lead about the incident cone is essential for stopping rays from material outside the field of view (Fig. 1*c*). The 5 inches of lead about the exit cone control rays from within the field of view that do not pass through the pinhole. Both cones contribute to the effectiveness of the pinhole, as pointed out in Figs. 1*a* and 1*b*.

Under conditions where a perfectly opaque material is not obtainable, the size of the pinhole is of no significance apart from its being the conjugation point of the two controlling cones. The ideal pinhole camera under those conditions, then, is achieved by the accurate alignment of two identical cones meeting at an *infinitesimal point*.

Some consideration must be given to the prevention of background fog caused by secondary radiations. The chief source of secondary radiation, it seems, is not from the region of the pinhole but from primary (gamma) effect on the atmosphere, metal lockers, *etc.*, surrounding the camera. Consequently, the primary source should be well shielded and/or the exit rays from



Pinhole camera refinements. (*a*)—Narrowing the incident and/or exit cones to a slot gives better control of the gamma rays in the dimension of the shorter arrow. (*b*)—Control can be obtained in the dimension of the slot by curving the slot, the degree of control being in proportion to the degree of curvature. (*c*)—The "slotting" and curvature can be obtained by a series of nesting cones focused on the pinhole as shown in cross section. For fusion of image, see text

the pinhole should be collimated to the emulsion.

The camera can be markedly improved by a modification of the Bucky diaphragm. In standard X-ray usage, this diaphragm is an alternating lamination of transmitting and non-transmitting material that is oriented on the radii from the target of the X-ray tube. It is placed under the patient, immediately over the negative and serves the function of stopping most of the divergent secondary rays produced in the patient's body. The filter is moved (Potter feature) during the exposure to prevent a striped effect on the negative.

The adaptation of the diaphragm to the pinhole camera would be to obtain better control of the gamma rays This can be visualized by Fig. 2 and the following steps.

If the exit cone (Fig. 2*a*) were flattened to a slot, extraneous rays not passing through the pinhole would be more quickly stopped in all planes except that of the slot.

Now if the slot were curved or fluted (Fig. 2*b*), the extraneous rays would be inhibited also in the plane of the slot, depending on the degree of curvature.

The slots could be multiplied and arranged in a radial fashion about the optical axis. The pinhole block would then need to be rotated about its optical axis to fuse the image formed on the negative. Much of the same effect could be obtained by nesting cones of transmitting and non-transmitting material (Fig. 2*c*). The walls of the cones would be of the same degree of thickness and the foci of all the cones would be the pinhole. To fuse the image the pinhole block would have to be moved in an eccentric fashion about the stationary pinhole, the degree of eccentricity being equal to the degree of wall thickness of one cone. Yet one other variant would be to use nested spiral cones, in which case fusion of image would be obtained by rotation on the axis of the summated cones.

Construction

To obtain an accurate alignment of the cones, the pinhole block was sand-cast in two sections with a steel chill forming the cones. The rough blocks were then gently swaged onto a similar cone turned down and still in a lathe. The lathe-turning of the blocks auto-

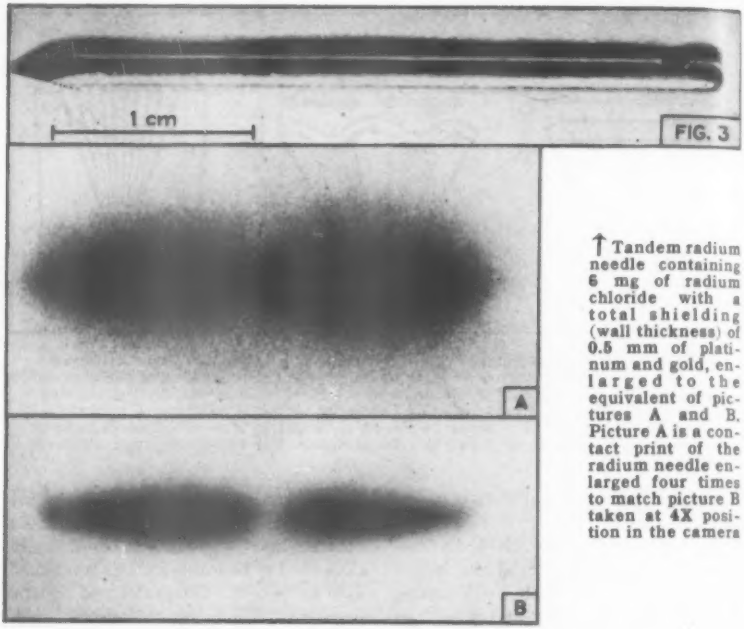


FIG. 3

↑ Tandem radium needle containing 6 mg of radium chloride with a total shielding (wall thickness) of 0.5 mm of platinum and gold, enlarged to the equivalent of pictures A and B. Picture A is a contact print of the radium needle enlarged four times to match picture B taken at 4X position in the camera

matically trued up the contact faces so that when the blocks were fastened together the axes of the cones accurately coincided.

To continue the slopes of the cones to a pinhole, two sheets of lead of calculated thickness were drilled with a tapered drill. They were then aligned with each other and fastened to the surface of one block. The other block was then aligned and fastened with bolts. Inspection was aided by use of a small mirror to reflect a focused beam of light down the throats of the cones. The pinholes were so small (*ca.* 0.1 mm) that transmitted light was of use only in aligning the two lead sheets.

The primary shield of lead was cast in two sections. The secondary or collimating shields were made of $\frac{3}{16}$-inch lead flashing which was cut and torch-welded into square pyramids. They were then cut and attached to removable mounts so that X-ray plate holders could be inserted at 1X, 2X, and 4X distances from the pinhole.

The frame work of the camera was constructed with attention to the alignment of all elements on the optical axis.

Application

The camera was tested with a 6 mg radium chloride needle. Exposures were made on ordinary X-ray film in cardboard holders inserted at the various levels. The lead backing was removed from the holders and simultaneous exposures were sometimes made at both 2X and 4X levels. The length of exposure necessary to obtain a good picture varied from about 24 hours at the 2X level to 48–72 hours at the 4X level. When the primary shields (approximately $2\frac{1}{2}$ inches of lead) and the secondary shields ($\frac{3}{16}$ inches) were used, there was no noticeable back-

ground fog. Without the shielding, a very noticeable fog is caused by secondary rays from the surroundings (in this case a metal locker). In an earlier camera designed by the authors, the wooden frame was assembled without nails or screws and a filter of tin, copper, and aluminum was used in front of the film. Such precautions are not necessary with the shielding just described.

The effectiveness of the design described here may be seen by referring to Fig. 3. Comparison of autographs and pinhole photographs of the needle show that the pinhole photographs give a sharper localization of the radium powder in the lumen of the needle. The critical importance of proximation of source and sensitive emulsion in autograph techniques is emphasized by the disparity between autographs and photographs caused by the 5 mm wall of the needle.

The resolution (*i.e.*, depiction of localization) can be improved by narrowing the two cones and sacrificing the field of view. The same end could be achieved and the field of view even enlarged by using a Bucky-type diaphragm. A carefully constructed diaphragm of this type would have the effect of assembling a large number of small cones into a summated, large cone, a sort of radial "collimation" with the pinhole as the center of the radii.

The camera was originally devised with the idea of applying it to biological tracer problems. That at the moment does not seem practical for two reasons. First, many of the tracers used in biological work have little gamma activity. Second, the exposure factor would limit work to tracers with a long half-life to compensate for the low specific activity required for biological tolerance. A possible solution to the latter point might be the use of a fluoroscopic screen plus Coltman's† electronic method of brightening the screen images.

* * *

The authors are indebted to Russel A. Peck of the Physics Department, Brown University, for information on gamma absorption in lead and to the Memorial Hospital of Pawtucket, R. I., for the use of their radium needles. Messrs. Sherwin and Benoit of the Browne & Sharpe Mfg. Co. solved the problem of casting the pinhole block free of defects.

† J. W. Coltman, *Radiology* **51**, 359 (1948).

BUT THERE ARE MORE IMPORTANT things for us to think about these days than our good public relations. There are more important considerations than our natural desire to win every battle. There is justice to think about, and freedom, and our integrity.

Justice and freedom have never been secure for very long in any one area of the world. None of us can say for sure what fate awaits them in the United States in the crisis through which we shall be going in the remainder of this century. Freedom and justice might survive this crisis; or they might not. They might perish and the efforts of scientists might be of little avail. What we scientists can do is to resolve that they shall not be allowed to perish without a fight. And those of us who do not wish to fight can at least refuse to help dig the grave.

—*Leo Szilard*, Bulletin of the Atomic Scientists **5**, *178 (1949)*

Effect of High-Voltage X-rays and Cathode Rays on Vitamins (Riboflavin and Carotene)

The results of tests using 3,000 kv radiations from a Trump generator to irradiate pure solutions of riboflavin and carotene are reported from a series of investigations with vitamins

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NO PUBLISHED REPORTS on the effects of ionizing radiations on riboflavin or carotene have come to the attention of the authors. A study of such effects would seem to be of value, since it has been shown that ionizing radiations may have potentialities as a means of food preservation (*3*, *14*). To evaluate the feasibility of using such radiations for the processing of foods, their effects on the important nutrients in foods must be determined. Among these nutrients are riboflavin and carotene.

Riboflavin is a component of one of the important enzyme systems of the body—the respiratory enzyme (Warburg's yellow enzyme). Carotene, one of the fat-soluble vitamins, a precursor of vitamin A, is not only important for growth and development but is also directly involved in the chemical changes upon which vision depends. Hence a study of the effects of ionizing radiations on riboflavin and carotene might have significance for those interested in research relating to cancer therapy by ionizing radiations and for those interested in problems of growth, development, and clearness of vision.

Reported below are the results obtained in experiments designed to determine the effects of cathode rays (electrons) and X-rays, electrostatically produced at 3,000 kilovolts, on pure solutions of riboflavin and carotene.

Equipment

A Trump generator (*14*, *16*, *17*) operating at 3,000 kv was used as a source of radiations. Cathode rays were obtained by allowing the electrons to impinge upon the sample located directly below the X-ray tube. X-rays were obtained by use of a gold, water-cooled target, one-fourth of an inch thick. The X-rays obtained were of low wavelength, the minimum being 0.004 A.U. The maximum intensity of radiations occurred at 0.008 A.U. The irradiation control panel of the Trump generator was equipped with an integrating roentgen-meter, which had been calibrated against Victoreen meters.

The quantities of ionizing radiations produced by the cathode rays were calculated from physical ionization measurements.‡ These quantities

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‡ The authors are grateful to Mr. Bernard Gregory of the High Voltage Laboratory, Dept. of Electrical Engineering, Massachusetts Institute of Technology, for determination of the physical ionization measurements from which the dosages of cathode rays were obtained.

(dosages) are expressed in terms of "roentgens-equivalent-physical" (rep), as described by Evans (*4*).

The electrons produced at 3,000 kv were more than sufficient to penetrate the samples irradiated in this study (*10*). However, as an additional check on the penetration of the ionizing radiations, use was made of oxidation-reduction dyes in an agar gel. Visual measurement of the depth of discoloration gave results that agreed well with physical measurements.

Samples were irradiated by cathode rays in small aluminum Petri dishes, with thin aluminum covers. The dishes were sealed with Scotch tape to prevent evaporation. Samples irradiated by X-rays were placed in small, stainless steel Petri dishes with stainless steel covers. These vessels have been previously described (*14*).

A Beckman quartz spectrophotometer, model DU, was used for determination of the ultraviolet absorption spectra.

Methods of Assay

Riboflavin was determined by fluorescence measurements. After irradiation, a 1-ml aliquot of the riboflavin sample was diluted to a known volume with a 20% pyridine solution in 2% acetic acid, the dilution depending on the amount of destruction of the riboflavin. This diluted aliquot was then placed in a cuvette, and its fluorescence was measured in a Pfaltz and Bauer fluorophotometer before and after reduction with sodium hydrosulfite. The quantity of riboflavin in the aliquot was measured by comparison of the fluorescence of the unknown with that of a control sample (standard U.S.P. riboflavin).

Carotene was measured by determining the extinction of duplicate 1-ml aliquots of each irradiated sample (diluted in petroleum ether) at 420 mμ in a Cenco Photelometer. The results were then compared with a standard curve representing the extinction of β-carotene (Smaco) in petroleum ether.

For the ultraviolet absorption spectra, readings were taken at every 5 mμ except at maxima and minima, where they were taken at intervals of one mμ.

Experimental

A 100-mg quantity of U.S.P. riboflavin was made to 1,000 ml in a volumetric flask with water acidified with three drops of glacial acetic acid. After the riboflavin was dissolved, the solution (in an amber-colored bottle) was kept in a mechanical refrigerator. This stock solution contained 100 micrograms (γ) of riboflavin per ml. Dilutions, when needed, were made with distilled water.

A stock solution of β-carotene (100 micrograms per ml) was prepared by dissolving 10 mg of β-carotene (Smaco) in petroleum ether and making to volume in a 100-ml volumetric flask. Further dilutions, when needed, were made from this stock solution with petroleum ether.

Five 2.5-ml samples of the stock solution of riboflavin were irradiated by

EDITOR'S NOTE: If cathode and X-radiations have potentialities as a means of processing foods, information is essential on how they affect vitamins, enzymes, and other nutrients in foodstuffs. Last year NUCLEONICS published the first paper on the subject. The vitamin reported on at that time was niacin (see reference *14* in the bibliography). The paper appearing here is the second report on the subject, by the same authors, with riboflavin and carotene as the nutrients under investigation. A third report, concerned with niacin, riboflavin, and ascorbic acid irradiated by soft X-rays, will be published next month.

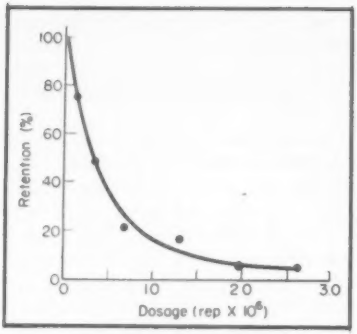


FIG. 1. Effect of 3,000-kv cathode rays on riboflavin (100 γ/ml)

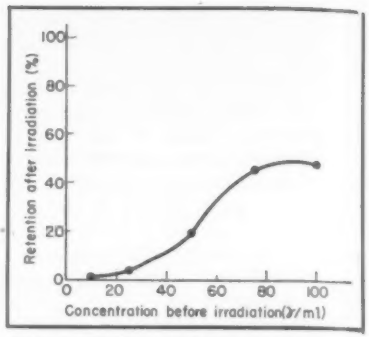


FIG. 2. Effect of 3,000-kv cathode rays on riboflavin of varying concentrations

cathode rays produced at 3,000 kv for dosages of from 0.33×10^6 to 2.64×10^6 rep (Table 1, upper half, and Fig. 1). Four 3-ml samples of the stock solution of β-carotene were irradiated by cathode rays produced at 3,000 kv for dosages of from 0.66×10^6 to 2.64×10^6 rep (Table 1, lower half, and Fig. 6).

Aliquots of the stock solution of riboflavin were diluted with distilled water to contain 75, 50, 25, and 10 micrograms of riboflavin per ml, and 2.5-ml samples of these dilutions were then irradiated by cathode rays produced at 3,000 kv for a dosage of 0.33×10^6 rep (Table 2, upper half, and Fig. 2).

Aliquots of the stock solution of carotene were diluted with petroleum ether to concentrations of 50 and 33.3 micrograms per ml, and 3-ml samples

TABLE 1

Effect of High-Voltage Cathode Rays on Stock Solutions of Riboflavin and Carotene

Concentration before irradiation..... 100 γ/ml
Volume irradiated..... 2.5–3 ml
Voltage..... 3 megavolts

Vitamin and Sample	*Time of Irradiation sec*	*Target Current μa*	*Dosage rep × 10⁶*	*Vitamin Retention after Irradiation %*
Riboflavin				
Control	..	..	....	100
Y	15	5	0.33	47.8
G	15	10	0.66	20.9
F	30	10	1.32	16.9
E	45	10	1.98	5.1
I	60	10	2.64	5.1
Carotene				
Control	..	..	....	100
Y	15	10	0.66	50
I	30	10	1.32	20
E	45	10	1.98	6
F	60	10	2.64	1

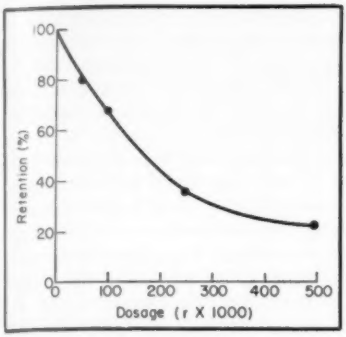


FIG. 3. **Effect of 3,000-kv X-rays on riboflavin (100 γ/ml)**

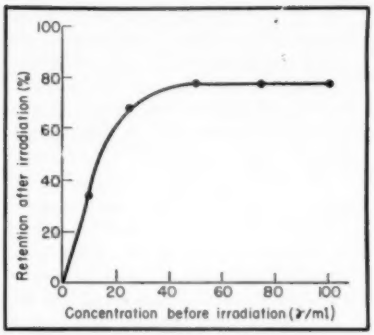


FIG. 4. **Effect of 3,000-kv X-rays on riboflavin of varying concentrations**

of these dilutions were irradiated by 3,000-kv cathode rays for dosages of 0.66×10^6 and 1.32×10^6 rep (Table 2, lower half, and Fig. 6).

Four 2.5-ml samples of the stock solution of riboflavin were irradiated by hard X-rays produced at 3,000 kv for dosages of from 50,000 to 500,000 roentgens (r) at a rate of approximately 500 r per sec (Table 3, upper half, and Fig. 3). Other riboflavin samples (2.5 ml) diluted to contain 75, 50, 25, and 10 micrograms of riboflavin per ml were irradiated by 3,000 kv X-rays for a dosage of 50,000 r (Table 3, lower half, and Fig. 4). Two samples with riboflavin in concentrations of 100 and 50 micrograms per ml, respectively, were irradiated by 3,000 kv X-rays for a dosage of 250,000 r (Table 3, lower half).

TABLE 2

Effect of High-Voltage Cathode Rays on Solutions of Riboflavin and Carotene of Varying Concentrations

Volume irradiated..... 2.5–3 ml
Voltage..... 3 megavolts

Vitamin and Sample	*Time of Irradiation sec*	*Target Current μa*	*Dosage rep × 10^6*	*Concentration of Vitamin*		*Retention of Vitamin %*
				Before Irrad. γ/ml	*After Irrad. γ/ml*	
Riboflavin						
Y	15	5	0.33	100	47.8	47.8
I	15	5	0.33	75	33.0	45.5
E	15	5	0.33	50	9.58	19.2
F	15	5	0.33	25	0.978	3.91
G	15	5	0.33	10	0.181	1.81
Carotene						
B	15	10	0.66	50	19	38
N	30	10	1.32	50	4	8
D	15	10	0.66	33.3	10	30
A	30	10	1.32	33.3	1.8	5.4

In an attempt to determine what changes occur in the riboflavin molecule when it is exposed to ionizing radiations, an aliquot of the stock solution of riboflavin was diluted to 50 micrograms per ml, and 2.5-ml samples of this diluted aliquot were irradiated for 50,000, 100,000, and 250,000 r. One-milliliter aliquots of the irradiated samples were then diluted to 5 ml with distilled water, and determinations of the ultraviolet absorption spectra were made with the spectrophotometer (Table 4 and Fig. 5).

Ultraviolet and visible absorption spectra were likewise determined on control and irradiated samples of carotene, to ascertain, if possible, which parts of the molecule were being modified by the irradiation.

Discussion of Results

Riboflavin in a concentration of 100 micrograms per ml was radiosensitive to ionizing radiations (cathode rays and X-rays produced at 3,000 kv), as shown by the data in Tables 1 and 3 and Figs. 1 and 3. Visual examination indicated that irradiation of the riboflavin was accompanied by a bleaching of the vitamin solution.

If ionizing radiations act upon riboflavin according to the "indirect action" theory (*13*), one might expect a greater destruction of riboflavin in more dilute solutions than in concentrated ones. Such was found to be the case, when irradiated by cathode rays and by X-rays produced at 3,000 kv (Tables 2 and 3). The same type of effect (indirect action) was observed by Forssberg (*5*) with catalase, by Dale (*2*) with carboxypeptidase, by the authors (*14*) with niacin, and by a number of other investigators with chemical compounds irradiated by ionizing radiations (*8*).

The relative radiosensitivities of niacin and riboflavin irradiated by both X-rays and cathode rays are compared

TABLE 3

Effect of High-Voltage X-Rays on Solutions of Riboflavin

Volume irradiated..... 2.5 ml
Voltage..... 3 megavolts

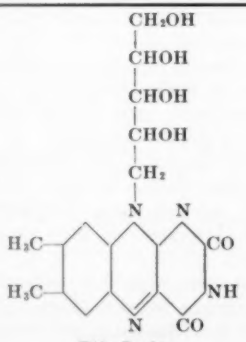
Sample No.	*Target Current μa*	*Time of Irradiation sec*	*Dosage*		*Concentration of Riboflavin*		*Retention of Riboflavin %*
			Total roentgens	*Rate r/sec*	*Before Irrad. γ/ml*	*After Irrad. γ/ml*	
Stock soln.							
4	110	122	50,000	410	100	79.1	79.1
17	80	205	100,000	488	100	67.8	67.8
24	100	468	250,000	534	100	35.2	35.2
28	100	887	500,000	563	100	22.4	22.4
30	Control	...	.....	...	100	100*	100*
Diluted							
4	100	78	50,000	641	100	77.8	77.8
15	100	78	50,000	641	75	58.4	77.9
24	90	85	50,000	589	50	39.4	77.7
28	90	89	50,000	561	25	17.0	68.1
17	100	87	50,000	575	10	3.3	33.0
30	100	498	250,000	501	50	7.6	15.2
24′	100	468	250,000	534	100	35.2	35.2

* Not irradiated

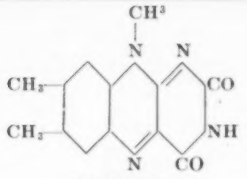
in Table 5. The niacin data are taken from previously reported experiments (*14*).* It is apparent that equal total dosages of irradiation of these two vitamins in solutions of the same initial concentration result in a more severe destruction of riboflavin than of niacin.

Riboflavin is chemically resistant to acids, bromine, and oxidizing agents such as hydrogen peroxide and concentrated nitric acid. Riboflavin possesses a relatively high degree of thermostability, and only slight destruction of this vitamin occurs when it is subjected to dry heat at 120° C for 6 hours (*7*). Hydrogen peroxide has been reported to be formed when any aqueous solutions are irradiated (*7*). Yet, chemically, oxidizing agents have no effect on riboflavin (*11*).

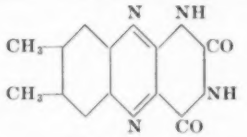
Riboflavin is sensitive to light. Irradiation by ultraviolet light or even by ordinary sunlight is destructive to riboflavin. According to Karrer (*11*), lumiflavin is formed in alkaline solutions of riboflavin in addition to a sugar compound that cannot be isolated as such, probably because of its rapid decomposition. In neutral or acid solution, the formation of lumichrome occurs (*12*):



Riboflavin

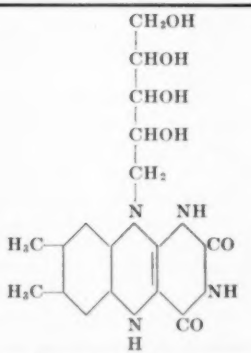


Lumiflavin



Lumichrome

Chemically, riboflavin is readily reduced to form leucolactoflavin or leucoflavin, a colorless compound (*11*):



Leucolactoflavin or **Leucoflavin**

Because riboflavin is extremely resistant to oxidation, ionizing radiations are unlikely to have an oxidative effect. In our experiments a few drops of acetic acid were added to the aqueous solution of riboflavin, bringing the *p*H down to 4.0. It is, therefore, improbable that lumiflavin was formed, for this compound is formed only in alkaline solutions.

Two possibilities, therefore, remain; first, that lumichrome is formed when

* In subsequent experiments (*9*), it has been found that niacin is decarboxylated long before the pyridine ring is split by cathode rays.

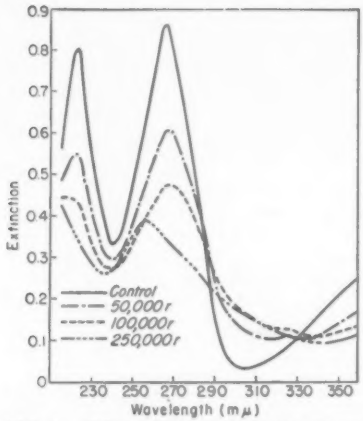


FIG. 5. Effect of high-voltage X-rays on the ultraviolet absorption spectrum of riboflavin

an acid or neutral solution of riboflavin is irradiated by ultraviolet light, and second, that leucoflavin is formed by chemical reduction.

The ultraviolet absorption spectra for pure riboflavin (not irradiated) shows a minor peak at a wavelength of 223 mμ and a major peak at a wavelength of about 267 mμ (Fig. 5). Karrer *et al.*(*12*) have shown that lumichrome has an absorption maximum at a wavelength of 255 mμ and a minimum at a wavelength of about 275 mμ. A secondary maximum occurs at 340 to 360 mμ. The peak at 255 mμ corresponds exactly to that obtained by irradiation of riboflavin with 250,000 r of X-rays (Table 4 and Fig. 5). This would seem to indicate that lumichrome may be formed by irradiation of riboflavin with X-rays.

The abrupt decline of the curve for the absorption spectrum of pure lumichrome after the peak at 255 mμ wavelength and the less abrupt decline of the curve for the sample of riboflavin irradiated by hard X-rays for a dosage of 250,000 r are possible indications that complete molecular disruption of a portion of the riboflavin molecules was taking place and that certain fragments of the irradiated riboflavin molecules were interfering. The fact that lumichrome has no peak at 223 mμ suggests that this compound was being formed when the riboflavin was irradiated.

This does not rule out the possibility that leucoflavin was being formed. It is known that ionizing radiations may cause oxidizable substances to be oxidized and reducible substances to be reduced (*1*). Hence it is possible that leucoflavin was being formed. The literature is devoid of references to the absorption spectra of this compound and of other derivatives of riboflavin, although it is remarkably complete in references to other chemical properties of these substances.

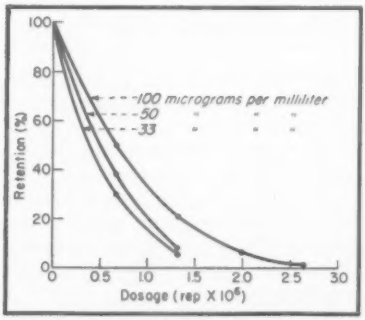


FIG. 6. Effect of 3,000-kv cathode rays on beta-carotene of varying concentrations

Carotene in petroleum ether in a concentration of 100 micrograms per ml was radiosensitive, 50% being destroyed by 0.66×10^6 rep (Table 1). This vitamin reacts to ionizing radiations in a manner similar to that shown by niacin, ascorbic acid, riboflavin, and chemical compounds that have been studied.

There was a greater destruction of the carotene in more dilute solutions (Table 2). When dry, pure β-carotene was irradiated by the maximum dosage of cathode rays used in this work (2.64×10^6 rep), there was a retention of 60% of the vitamin. Comparison

TABLE 4

Effect of High-Voltage X-Rays on Ultraviolet Absorption Spectra of Pure Solutions of Riboflavin

Concentration before irradiation..... 50 γ/ml
Volume irradiated..... 2.5 ml
Voltage..... 3 megavolts
Samples diluted 1:5 for U.V. spectra

Sample No.	*Target Current μa*	*Time of Irradiation sec*	*Dosage*		*Absorption Spectra*		*Extinction Coefficient**
			Total roentgens	*Rate r/sec*	*Maxima mμ*	*Minima mμ*	
Control	...	...	.....	...	223	240	80.4
					267	304	86.1
1	50	98	50,000	510	223	240	55.0
					267	...	60.9
6	50	211	100,000	484	248	...	43.4
					267–8	241	47.9
30	100	498	250,000	501	256	237	38.8

* At maximum absorption wavelength

of this value with the data in Tables 1 and 2 gives additional evidence of the effect of dilution. This comparison would seem to indicate that the radiations had an indirect action on the solute and that the carotene reacted with the irradiation products.

Ordinarily the unknown irradiation products are thought of as "activated water." In this case, however, other compounds were presumably formed, because an organic solvent was used. Yet similar phenomena apparently took place. The question may be asked whether the cathode rays affected the solvent (petroleum ether), which then in turn affected the solute (β-carotene). To answer this question, petroleum ether alone was irradiated by 2.64×10^6 rep of cathode rays, and subsequently β-carotene was added to the irradiated solvent. In this case there was no destruction of the vitamin. This indicates that whatever destruction of β-carotene was caused by action of the cathode rays on the solvent took place at the time of irradiation.

It was impossible to determine from the ultraviolet and visible absorption spectra of the irradiated carotene which

TABLE 5

Relative Radiosensitivities of Niacin and Riboflavin

Type of Irradiation and Total Dosage	*Vitamin*	*Concentration before Irradiation γ/ml*	*Retention after Irradiation %*
X-Rays:	Niacin	50	43
250,000 roentgens	Riboflavin	50	15.2
Cathode Rays:	Niacin	100	94
660,000 rep	Riboflavin	100	20.9

part of the molecule was modified by the irradiation. These spectra merely showed a complete shattering of the molecule with a low extinction at a wavelength of 452 mμ.

Summary and Conclusions

1. The effects of high-voltage, electrostatically produced X-rays and cathode rays (electrons) on pure solutions of riboflavin and the effects of such cathode rays on pure solutions of carotene were determined.

2. Riboflavin was radiosensitive to both cathode rays and X-rays.

3. Carotene was radiosensitive to cathode rays, 50% of a petroleum ether solution containing 100 micrograms of carotene per milliliter being destroyed by 0.66×10^6 rep.

4. The destruction of both riboflavin and carotene was greater, the more dilute the solutions. This is indicative of an "indirect action" of the ionizing radiations upon these two vitamins. Since the carotene was irradiated in a solution of petroleum ether, the activated water hypothesis cannot be used to explain this finding. Presumably, other compounds were formed by the irradiation of the petroleum ether.

5. From spectroscopic evidence it would appear that lumichrome may be formed when riboflavin is irradiated by high-voltage cathode rays. There is evidence also that some of the riboflavin molecules are broken down into smaller fragments.

* * *

This report is the second of a series regarding irradiation studies conducted by the staff of the Department of Food Technology, MIT, with the assistance of grants-in-aid from the following organizations: American Can Co., Maywood, Ill.; Dow Chemical Co., Midland, Mich.; Nestlé Co., New York, N. Y.; Pillsbury Mills, Inc., Minneapolis, Minn.; Standard Brands, Inc., New York, N. Y.; Wilson & Co., Chicago, Ill.

Grateful acknowledgment is made to Professor John G. Trump, the late Mr. Arthur M. Clarke, and Mr. Kenneth A. Wright, of the Department of Electrical Engineering, MIT, for their cooperation in placing the Trump generator at the disposal of the authors.

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Introduction to the Theory of Diffusion and Slowing Down of Neutrons—IV*

The subjects of improvements on age theory and slowing-down density are taken up in concluding this series of papers which discussed the diffusion theory, improvements on it, and derivation of the age theory.

By R. E. MARSHAK,† H. BROOKS,‡ and H. HURWITZ, Jr.‡

Improvements on Age Theory

WE HAVE SEEN that the age equation in the case of no absorption may be written

$$\nabla^2 q = \frac{\partial q}{\partial \tau} - S(\mathbf{r})\delta(\tau) \qquad (147)$$

where $S(\mathbf{r})$ is the rate at which neutrons are produced per unit volume at the point $\mathbf{r}$, τ is the age as defined by Eq. 136, and q is the slowing-down density. We are assuming that all neutrons are produced at the same energy, which corresponds to $\tau = 0$. The slowing-down density q is defined by

$$q(\mathbf{r},\tau) = \xi N_0(\mathbf{r},u)v(u)\sigma(u)$$

where $N_0(\mathbf{r},u)$ is the number of neutrons per unit volume and per unit logarithmic energy range. The quantity $N_0(\mathbf{r},u)v(u)\sigma(u)du$ is the rate at which neutrons in the range du are suffering collisions per unit volume. Since the average change in u per collision is ξ, it is clear that, within the limits of validity of age theory, $q(\mathbf{r},\tau)$ may be regarded as the "current density in u space," *i.e.*, the rate per unit volume at which neutrons are passing across the energy value corresponding to τ. Since we have assumed no absorption, conservation of neutrons requires that the integral of $q(\mathbf{r},\tau)$ over all space be equal to the rate at which neutrons are being produced, no matter what τ is. That this condition is actually contained in Eq. 147 can be seen by integrating Eq. 147 over all space. Since the integral of, $\nabla^2 q$ over all space is zero, we find

$$\frac{\partial}{\partial \tau}\int q(\mathbf{r},\tau)dV = \delta(\tau)\int S(\mathbf{r})dV \qquad (148)$$

Hence

$$\begin{aligned} \int q(\mathbf{r},\tau)dV &= 0 \qquad \tau < 0 \\ &= \int S(\mathbf{r})dV \qquad \tau > 0 \end{aligned} \qquad (149)$$

Equation 149 is strictly true only in the age approximation of slowing-down theory (*cf.* Eq. 192 and *ff.*)

It was shown in Part III that the solution of Eq. 147 for a plane source at $z = 0$ is:

$$q(z,\tau) = \frac{e^{-\frac{z^2}{4\tau}}}{\sqrt{4\pi\tau}} \qquad (150)$$

We can find the point source solution by means of the relation in Eq. 27.

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* This is the last of a series of papers compiled from notes taken by H. Brooks and H. Hurwitz, Jr., on lectures given at the General Electric Company by R. E. Marshak during the summer of 1946. A large part of the work described was performed under the auspices of the Atomic Energy Project and thus will be presented in volumes of the National Nuclear Energy Series (Manhattan Project Technical Section).

See NUCLEONICS **4**, No. 5, 10; No. 6, 43; and **5**, No. 1, 53 (1949) for preceding articles in this series.

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It is:

$$q(r,\tau) = \frac{e^{-\frac{r^2}{4\tau}}}{(4\pi\tau)^{3/2}} \qquad (151)$$

The average values of z^2 and r^2 can be found from Eqs. 150 and 151. They are:

$$\overline{z^2} = 2\tau$$
$$\overline{r^2} = 3\overline{z^2} = 6\tau \qquad (152)$$

The quantity $\sqrt{\tau}$ has been referred to as the slowing-down length L_s. We therefore have:

$$L_s^2 = \frac{\overline{r^2}}{6} = \frac{\overline{z^2}}{2} \qquad (153)$$

If the medium in which the neutrons are produced is not infinite in extent, the simple solution in Eq. 151 does not apply. In first approximation, the boundary condition to be applied to $q(\mathbf{r},\tau)$ is that it vanish at the surface of the medium. (This is equivalent to the simple diffusion-theory boundary condition that the neutron density should vanish on all surfaces. See below for a more accurate discussion of the boundary conditions at a surface.) The problem to be solved is then equivalent to the time-dependent heat flow problem in a region whose surface is maintained at temperature zero. It can be solved for a large class of regions with different distributions of sources. The results for many cases of practical importance are given by Wallace and LeCaine (*2*).

An interesting problem which can be treated by age theory (*27*) is that of a point source in a semi-infinite medium bounded by another medium with the same atomic scattering cross section as a function of energy, but different density (see Fig. 8).

This means that:

$$\frac{\lambda_1(E)}{\lambda_2(E)} = \frac{\rho_2}{\rho_1} \qquad (154)$$

so that for a particular value of neutron energy

$$\frac{\tau_1}{\tau_2} = \frac{\rho_2^2}{\rho_1^2} = D \qquad (155)$$

The age equations in the two media are:

$$\nabla^2 q_1 = \frac{\partial q_1}{\partial \tau_1} - \delta(\tau)\delta(\mathbf{r}_0)$$
$$\nabla^2 q_2 = \frac{\partial q_2}{\partial \tau_2} \qquad (156)$$

It is convenient to consider q_2 as a function of τ_1 so that equal arguments refer to equal neutron energies in the two regions. Then we have:

$$\nabla^2 q_2 = D\frac{\partial q_2}{\partial \tau_1} \qquad (157)$$

The boundary conditions to be applied at the surface are

$$q_2 = \frac{\lambda_1}{\lambda_2} q_1 = \sqrt{D}\, q_1$$

$$\frac{\partial q_2}{\partial z}\Big|_{z=0} = \left(\frac{\lambda_1}{\lambda_2}\right)^2 \frac{\partial q_1}{\partial z}\Big|_{z=0} = \sqrt{D}\,\frac{\partial q_1}{\partial z}\Big|_{z=0} \qquad (158)$$

(The latter relation comes from the continuity of current at a given age.) The problem can be treated by the method of Laplace transforms. If ϕ_1 and ϕ_2 are the Laplace transforms of q_1 and q_2, respectively, with respect to the argument τ_1 (*cf.* Eq. 139), then Eqs. 156 and 157 are:

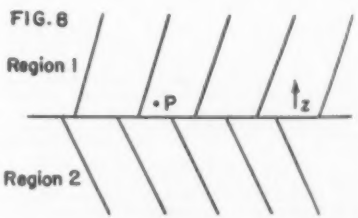
$$\nabla^2\phi_1(\eta) = \eta\phi_1(\eta) - \delta(\mathbf{r}_0)$$
$$\nabla^2\phi_2(\eta) = D\eta\phi_2(\eta) \qquad (159)$$

The boundary condition in Eq. 158 becomes:

$$\phi_2 = \sqrt{D}\,\phi_1$$
$$\frac{\partial\phi_2}{\partial z}\Big|_{z=0} = D\frac{\partial\phi_1}{\partial z}\Big|_{z=0} \qquad (160)$$

Equations 159 and 160 correspond to the problem of a point source of sound waves in a semi-infinite medium bounded by a second medium of different accoustical impedance (Sommerfeld

FIG. 8



antenna problem). A formal solution to this problem can be obtained, so that to find q_1 and q_2 one simply has to take the Laplace inverse with respect to η of the solution to the antennae problem with η as a parameter. In practice, the necessity of taking a Laplace inverse is sometimes a help rather than a hindrance because, after the Laplace inverse has been taken, the solutions sometimes turn out simpler than before the inverse has been taken. For example, if the source of neutrons lies on the interface, one can obtain a closed expression for $q_1(\mathbf{r},\tau)$ at the interface. The expression is

$$q(\mathbf{r},\tau) = \frac{1}{(4\pi\tau)^{3/2}} \frac{1}{\left(1 - \frac{1}{D}\right)} \left[E_i\left(-\frac{r^2 D}{4\tau}\right) - E_i\left(\frac{-r^2}{4\tau}\right) \right] \quad (161)$$

where $-E_i(-\chi) = \int_\chi^\infty \frac{e^{-u}}{u}\, du.$

Solution Near a Boundary

We have seen that in one-velocity problems the neutron density is not exactly zero at the boundary between a scattering medium and a vacuum. The same statement holds for many-velocity problems. In order to obtain an idea of boundary effects in slowing-down problems, the problem of a plane source of neutrons near a plane boundary (*cf.* Fig. 5 in Part I) has been investigated in some detail. The simplest way of treating this problem is to retain only the first two terms in the expansion of the neutron distribution function in spherical harmonics. This approximation leads to the simple age equation throughout the medium and the boundary condition (as shown in Part I) that q vanish at a distance $\frac{2}{3}\lambda_t$ beyond the surface. (In Part I, the scattering was assumed isotropic, but if the same simple argument is carried through for non-isotropic scattering the extrapolated endpoint turns out to be $\frac{2}{3}\lambda_t$.) We therefore impose on q the boundary condition at the surface:

$$\frac{3}{2} q - \frac{\lambda_t(\tau)}{3} \int \frac{\partial q}{\partial z} = 0 \quad \text{at} \quad z = 0 \quad (162)$$

The mathematical problem to be solved is then equivalent to the corresponding heat problem where the surface radiates into a medium at temperature zero (*cf.* Carslaw*). If $\lambda_t(\tau)$ is constant, the solution is:

$$q = \left\{ \frac{e^{-\frac{(z-z')^2}{4\tau}}}{\sqrt{4\pi\tau}} + \frac{e^{-\frac{(z+z')^2}{4\tau}}}{\sqrt{4\pi\tau}} - \frac{3}{2\lambda_t} e^{\frac{9\tau}{4\lambda_t^2} + \frac{3(z+z')}{2\lambda_t}} \left[1 - \operatorname{erf}\left(\frac{3\sqrt{\tau}}{2\lambda_t} + \frac{z+z'}{2\sqrt{\tau}} \right) \right] \right\} \quad (163)$$

where $\operatorname{erf}(\chi) = \frac{2}{\sqrt{\pi}} \int_0^\chi e^{-y^2} dy \quad (164)$

For large τ and not too large z, the solution in Eq. 163 becomes

$$q = \left\{ \frac{e^{-\frac{(z-z')^2}{4\tau}}}{\sqrt{4\pi\tau}} - \frac{e^{-\frac{(z+z'+\frac{4}{3}\lambda_t)^2}{4\tau}}}{\sqrt{4\pi\tau}} \right\} \quad (165)$$

This is what one would obtain by applying the boundary condition $q = 0$ at $z = -\frac{2}{3}\lambda_t$ instead of the condition represented in Eq. 162. Thus the effective extrapolated endpoint is related to λ_t, and not to the slowing-down length $\sqrt{\tau}$. Some information can be obtained in the case of variable mean free path by assuming a variation of the form

$$\lambda_t(\tau) = \lambda_t(0) + A e^{-\alpha\tau}, \quad (166)$$

If A is assumed to be small, one can treat the term $A e^{-\alpha\tau}$ as a perturbation. It then turns out that the extrapolated endpoint corresponding to a particular age τ depends primarily on the value of $\lambda_t(\tau)$ for that age. Since λ_t usually decreases for lower energy (larger age), the effective extrapolated endpoint for large age may be very small.

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* H. W. Carslaw, "Mathematical Theory of the Conduction of Heat in Solids," 2d edition, page 173 (Dover Publications, New York, 1945).

Improvements on Slowing-down Density

A natural way to improve age theory is to keep more terms in the expansion of $\Psi(z,u,\mu)$ in spherical harmonics (*cf.* Part III) and to keep higher derivatives of Ψ with respect to u (*cf.* Eq. 127). But instead of introducing the higher derivatives of Ψ it is more convenient to use the method of Laplace transforms. To illustrate this method of treatment let us consider the case in which $\lambda(u) = 1/[\sigma(u)]$ is independent of u and the source is a plane δ-source. The fundamental equation in an infinite medium is then (*cf.* Eq. 121):

$$\mu\lambda\frac{\partial\Psi}{\partial z} = -\Psi + \int du' \int d\Omega'\Psi(z,\mu',u')f(\mu_0,u-u') + \frac{\delta(z)\delta(u)}{4\pi} \quad (167)$$

If we make a Laplace transform of this equation (*i.e.*, multiply by $e^{-\eta u}$ and integrate from 0 to ∞), we obtain (assuming $\lambda = 1$):

$$\mu\frac{\partial\phi(z,\mu,\eta)}{\partial z} = -\phi(z,\mu,\eta) + \int d\Omega'\phi(z,\mu',\eta)g(\mu_0,\eta) + \frac{\delta(z)}{4\pi} \quad (168)$$

where $\phi(z,\mu,\eta)$ is the Laplace transform of $\Psi(z,\mu,u)$ and $g(\mu_0,\eta)$ is the Laplace transform of $f(\mu_0,u)$. We have made use of the convolution theorem for Laplace transforms which is:

$$\mathfrak{L}\int_0^u F_1(u')F_2(u-u')\,du' = \mathfrak{L}F_1\,\mathfrak{L}F_2 \quad (169)$$

(The symbol $\mathfrak{L}F$ indicates the Laplace transform of F.) Equation 168 shows that the slowing-down problem is equivalent to an infinite set of one-velocity problems with η as a parameter. Hence, to solve the slowing-down problem, one must solve the one-velocity problems and then take the Laplace inverse. The one-velocity problems may be treated by means of the spherical harmonic method. For example, in the P_2 approximation, Eq. 168 becomes (*cf.* Eq. 79):

$$\frac{\partial\phi_1}{\partial z} = -\phi_0 + \phi_0 g_0 + \delta(z) \quad (170)$$

$$\frac{1}{3}\frac{\partial\phi_0}{\partial z} + \frac{2}{3}\frac{\partial\phi_2}{\partial z} = -\phi_1 + \phi_1 g_1$$

$$\frac{2}{5}\frac{\partial\phi_1}{\partial z} = -\phi_2 + \phi_2 g_2$$

To solve Eq. 170, one makes a Fourier transform of the equations with respect to z. If we write:

$$\chi_s(y,\eta) = \int_{-\infty}^{+\infty} e^{-iyz}\phi_s(z,\eta)dz \quad (171)$$

Eq. 170 becomes:

$$iy\chi_1 = -\chi_0 + \chi_0 g_0 + 1$$

$$\tfrac{1}{3}iy\chi_0 + \tfrac{2}{3}iy\chi_2 = -\chi_1 + \chi_1 g_1 \quad (172)$$

$$\tfrac{2}{5}iy\chi_1 = -\chi_2 + \chi_2 g_2$$

These equations may then be solved for $\chi_0(y,\eta)$ so that in order to obtain the neutron density as a function of z and u, one has only to perform Laplace and Fourier inversions.

In order to arrive at practically useful results, one need determine the neutron density only for large values of u. In that case, taking the Laplace inverse is facilitated by the use of Tauberian theorems (*cf.* Doetsch's book* on Laplace transforms). These theorems state that in order to find the behavior of a function for large values of the argument, one must study the Laplace transform of the function for small values of the transform variable. Therefore, the functions g_i in Eq. 172 can be expanded in powers of η, and we find:

$$g_0 = 1 - \xi\eta + \cdots$$
$$g_1 = \frac{2}{3M} + \cdots \quad (173)$$

The number of powers of η which are to be retained depends on the number of spherical harmonics which are retained, *e.g.*, in the P_1-approximation which corresponds to age theory, one keeps the powers of η indicated in Eq. 173. Note that one less power of

* G. Doetsch, "Theorie und Anwendung der Laplacetransformation" (Springer, Berlin, 1937).

η is retained in each successively higher g_k. In solving Eq. 172 for $\chi_0(y,\eta)$, higher and higher powers of y^2 enter as more spherical harmonics are included.

The slowing-down problem has been treated by the above method, and corrections to the Gaussian distribution have been obtained. In the P_3-approximation, the result is (*26*):

$$q(z,\tau) = \frac{e^{-\frac{z^2}{4\tau}}}{\sqrt{4\pi\tau}}\left(1 + \frac{1}{4u}Q_1 + \cdots\right) \quad (174)$$

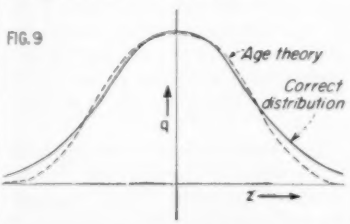
$$Q_1 = \left[(A_{11} - 2A_{10}) - 2(2A_{11} - A_{10})\frac{z^2}{2\tau} + A_{11}\frac{z^4}{4\tau^2}\right]$$

$$\left.\begin{aligned} A_{11} &= \frac{11}{15} \\ A_{10} &= -\frac{1}{15} \end{aligned}\right\} M = 1$$

$$\left.\begin{aligned} A_{11} &= \frac{18}{5M}\left(1 - \frac{2}{3M} + \cdots\right) \\ A_{10} &= -\frac{4}{15M}\left(1 - \frac{7}{3M} + \cdots\right) \end{aligned}\right\} M \gg 1$$

The corrections to q as given by Eq. 174 have the effect of making the distribution broader, *i.e.*, increasing the tails of the distribution. See Fig. 9.

FIG. 9



The procedure outlined above for finding q is essentially equivalent to the calculation of q by the method of moments.* If the average values of z^{2n} are known for all n as a function of τ, then $q(z,\tau)$ can be found by the well-known relationship between a distribution function and its moments. From Eq. 171:

$$\overline{z^{2n}}(\eta) = \int_{-\infty}^{+\infty} z^{2n}\phi_0(z,\eta)dz = i^{2n}\frac{d^{2n}}{dy^{2n}}\chi_0(y,\eta)\Big]_{y=0} \quad (175)$$

The Laplace transforms of the moments are therefore related to the values of $\chi(y,\eta)$ near $y = 0$. The moments may thus be obtained for large u by making expansions in powers of y^2 and η (keeping equal powers of y^2 and η). For slowing down by a single element with $\lambda(u)$ not necessarily constant, a closed expression may be obtained for $\overline{z^2}$ [*cf.* (*3*)].

Expansions of $q(z,\tau)$ of the type given in Eq. 174 cannot be expected to give accurate results when the distance expressed in units of the mean free path is not small compared to u/ξ, *i.e.*, when $z\lambda/3\tau$ is not much less than one. (The quantity u/ξ is a measure of the number of collisions required to change the logarithm of neutron energy by an amount u.) The Gaussian-type solution of Eq. 174 should only be a good approximation when the total distance traveled by a neutron [$\sim \lambda(u/\xi)$] is large compared to z. In fact, for extremely large z, the neutron density should fall off like $e^{-z/\lambda}$ rather than $e^{-z^2/4\tau}$ since neutrons have a probability of order $e^{-z/\lambda}$ of reaching the distance z without being scattered at all.

The problem of finding how the neutron distribution goes over from the Gaussian form at small distances to the exponential form at large distances has recently received much attention. Quite similar methods of treatment have been used by Wick, Bethe, and others.†

* The method of moments is, in fact, the one by which Placsek first obtained the result given in Eq. 174.

† M. Verde, G. C. Wick, *Phys. Rev.* **71**, 852 (1947).
G. C. Wick, *Phys. Rev.* **75**, 738 (1949).
H. A. Bethe, Declassified Report, KAPL-56.
H. A. Bethe, L. Tonks, H. Hurwitz, Jr. (to be published in *Phys. Rev.*).
I. Waller, *Arkiv Mat. Astron. Fysik* **34A**, No. 5 (1946).

The procedure is to make use of the one-velocity form of the problem (Eq. 168) which is obtained by making a Laplace transform with respect to u of the Boltzmann equation. For a given η, Eq. 168 can be solved by making a Fourier transform with respect to z as described in Part I. For large z the solution will behave like $Ae^{-\nu z}$ where both A and ν are functions of η. If this asymptotic form of the one-velocity solution is used, the slowing-down density, which is the Laplace inverse of the one-velocity solution, is given by a relation of the form

$$q = \frac{1}{2\pi i}\int_{-i\infty}^{+i\infty} d\eta A(\eta) e^{-\nu(\eta)z + u\eta} \quad (176)$$

The integral with respect to η can be performed by the method of steepest descents. The saddle point is determined by the condition that the derivative with respect to η of the exponent of the integrand is zero, *i.e.*,

$$\frac{d\nu}{d\eta} = \frac{u}{z} \quad (177)$$

This condition determines η, and hence ν as a function of (z/u). The exponent at the saddle point is of the form: $-u(-\eta + \nu z/u) = -u\varphi(z/u)$. The second derivative of the exponent at the saddle point is of the form u times a function of z/u so that q can be written

$$q = \frac{K(z/u)}{\sqrt{u}} e^{-u\varphi(z/u)} \quad (178)$$

For small z/u, $\varphi \sim z^2/u^2$, giving the age theory result for q, whereas for large z/u, $\varphi \sim z/u$, giving the exponential behavior.* The calculation of $\varphi(z/u)$ requires a knowledge of ν as a function of η, and this must be obtained numerically for intermediate values of z/u.

Wick and his collaborators did most of their numerical work for the case of hydrogen ($M = 1$), whereas Bethe and his collaborators slanted their numerical work toward scatterers with larger mass.

As was shown in Part I, the asymptotic decay constant ν in a one-velocity problem is determined by the location of the pole in the Fourier transform of the one-velocity solution. The Fourier transform of the one-velocity problem (Eq. 168) has the form (*cf.* Eq. 172):

$$(1 + iy\mu)\chi(y,\eta,\mu) = \int d\Omega \chi(y,\eta,\mu')g(\mu_0,\eta) + \frac{1}{4\pi} \quad (179)$$

At the pole, *i.e.*, when $iy = \nu(\eta)$, the homogeneous equation obtained by omitting the source term $1/(4\pi)$ must have a non-vanishing solution. In finding the eigenvalue of the homogeneous equation the spherical harmonic method converges slowly when z is not small compared to $\lambda u/\xi$ since the neutron distribution is far from isotropic.

For $M > 1$ it is, however, a good approximation in the practical range to keep only two or three terms in the expansion of the collision function transform $g(\mu_0,\eta)$ in spherical harmonics, and this enables the eigenvalue of the homogeneous equation to be calculated without undue labor.† For $M = 1$ the comparatively simple form of $g(\mu_0,\eta)$ allows the calculations to be performed with more terms retained in the spherical harmonic expansion of $g(\mu_0,\eta)$.

For extremely large values of z, where many terms in the equation of $g(\mu_0,\eta)$ must be retained, Wick developed ingenious methods for calculating the eigenvalue which are based on the fact that most of the neutrons at large z are moving in very nearly the forward direction.

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* For extremely large z/u, Wick has shown that $\varphi \sim z/u - A(z/u)^{\frac{1}{2}} + B$. Thus the neutron density does not fall off with distance quite as rapidly as the unscattered neutrons.

† From Eq. 172 it would appear that as higher and higher powers of iy are retained, more and more of the coefficients g_i should be included. This is not true in practice, however, since the higher g_i's have very small numerical effect on the eigenvalue in the interesting range.

The case of variable mean free path is mathematically more complicated than the constant-mean-free-path case. From the physical point of view it is, however, somewhat simpler, since the penetration to large distances is due primarily to the neutrons of highest energy which have the largest mean free path.

Wick and collaborators studied the variable-mean-free-path case with simplified assumptions about the behavior of the scattering cross section as a function of energy. It can be shown quite generally that at large distances the neutron density behaves as $z^{\delta}e^{-z/\lambda}$ where λ is the mean free path for the energy corresponding to that of the source, and δ is a constant which depends on how rapidly the mean free path decreases with decreasing energy.

Slowing Down with Capture (*28*)

If the neutron capture cross section is small and a slowly varying function of energy, it may be taken into account within the framework of age theory. If $h(u)$ represents the ratio of scattering to total cross section

$$h(u) = \frac{\sigma_s(u)}{\sigma(u)} \qquad (180)$$

the fundamental slowing-down equation becomes

$$\lambda(u)v \cdot \text{grad}\ \Psi(\mathbf{r},u,\mu) = -\Psi(\mathbf{r},u,\mu) + \int_0^u du' \int d\Omega' f(\mu_0, u-u') \times h(u')\Psi(\mathbf{r},u',\mu') + \frac{S(\mathbf{r},u)}{4\pi} \qquad (181)$$

where

$$\Psi = N(\mathbf{r},\mu,u)\sigma(u)v(u) \qquad (182)$$

Following the procedure outlined in Part III, we find from Eq. 181:

$$-\frac{1}{\sigma^2 3[1-h(u)\overline{\cos\theta}]}\nabla^2\Psi_0(\mathbf{r},u) = -[1-h(u)]\Psi_0(\mathbf{r},u) - \xi\frac{\partial h(u)\Psi_0(\mathbf{r},u)}{\partial u} + S(\mathbf{r},u) \qquad (183)$$

Hence, defining

$$q = \sigma_s(u)v(u)N_0(\mathbf{r},u)\xi = h\Psi_0 \qquad (184)$$

we obtain (since h is close to one):

$$-\frac{\lambda^2}{\left(1-\frac{2}{3M}\right)}\nabla^2 q = -\frac{\sigma-\sigma_s}{\sigma}q - \xi\frac{\partial q}{\partial u} + \xi S(\mathbf{r},u) \qquad (185)$$

Eq. 185 is often written in the equivalent form

$$\partial\xi\sigma_s N_0 v/\partial u = S(\mathbf{r},u) - \sigma_a N_0 v + \nabla\cdot\frac{1}{3\sigma_{tr}}\nabla N_0 v \qquad (186)$$

where $\sigma_{tr} = \sigma_s(1-\overline{\cos\theta}) + \sigma_a$

Here σ_a is the absorption cross section (in cm^{-1}) and $\overline{\cos\theta}$ is the average value of the cosine of the scattering angle. If more than one type of scatterer is present, ξ must be averaged over the different scatterers, each being weighted according to its scattering cross section. The terms in Eq. 186 have the following physical significance. The left-hand side is the rate at which slowing-down density changes with increasing u. The right-hand side is the rate at which neutrons are produced per unit volume and unit range in u by the source minus the rate at which neutrons are being absorbed per unit volume and range in u, plus the rate at which neutrons are diffusing into a unit volume element from the surrounding region.

If we define

$$\tau = \int_0^u du' \frac{\lambda^2(u')}{\xi\left(1-\frac{2}{3M}\right)} \qquad (187)$$

as in Eq. 136, we find from Eq. 185:

$$-\frac{\partial q}{\partial\tau} + \nabla^2 q = \frac{(\sigma-\sigma_s)}{\lambda}\left(1-\frac{2}{3M}\right)q - \delta(\tau)Q(\mathbf{r}) \qquad (188)$$

where we have taken for $S(\mathbf{r},u)$ a delta function in u. Equation 188 can be simplified by letting

$$q = q'\exp\left[-\int_0^\tau \frac{(\sigma-\sigma_s)}{\lambda}\left(1-\frac{2}{3M}\right)d\tau'\right] = q'\exp\left[-\int_0^u \frac{(\sigma-\sigma_s)}{\xi\sigma}du'\right] \qquad (189)$$

Then Eq. 188 becomes:

$$-\frac{\partial q'}{\partial \tau} + \nabla^2 q' = -\delta(\tau)Q(\mathbf{r}) \quad (190)$$

which is of the same form as the equation without absorption. Hence in the age theory approximation, the effect of absorption can be factored out. The solution is equal to the solution for no absorption times the factor

$$H(\tau) = \exp\left[-\int_0^\tau \frac{(\sigma - \sigma_a)}{\lambda}\left(1 - \frac{2}{3M}\right)d\tau'\right] \quad (191)$$

$$= \exp\left(-\int_0^u \frac{\sigma_a}{\xi\sigma_s}\,du'\right)$$

When narrow but strong absorption resonances occur, it is more accurate to replace the ratio σ_a/σ_s under the integral in the exponent by $\sigma_a\ (\sigma_a + \sigma_s)$ as was done by Wigner (*cf.* Eq. 213). Note that when absorption occurs the integral of slowing-down density over all space is not independent of τ, but decreases with increasing τ according to the factor $H(\tau)$ (*cf.* Eq. 149). In problems where more than one medium must be considered, the effect of absorption cannot be factored out in this simple way unless $\sigma_a/(\xi\sigma_s)$ is the same in all the media.

To find the effect of capture on the neutron spectrum more accurately, it is worthwhile to examine the space independent problem. If Eq. 181 is integrated over $\mathbf{r}$ and $\mathbf{\Omega}$, it becomes:

$$\overline{\mathbf{\Psi}}_0(u) = \int_0^u du'\,\overline{\mathbf{\Psi}}_0(u')f_0(u - u')h(u') + \delta(u) \quad (192)$$

where

$$\left.\begin{aligned} f_0(u) &= \frac{(M-1)^2}{4M}\,e^{-u} \qquad 0 < u < \epsilon \\ &= 0 \qquad\qquad\qquad u > \epsilon \end{aligned}\right\}$$

$$\epsilon = \ln\left(\frac{M+1}{M-1}\right)^2$$

$$\overline{\mathbf{\Psi}}_0(u) = \int_{-\infty}^{+\infty} d\mathbf{r}\mathbf{\Psi}_0(\mathbf{r})$$

(We have assumed a δ-function source.) Let us first treat Eq. 192 under the assumption that $h(u) = 1$. If we make a Laplace transform and use the convolution theorem we obtain:

$$Z(\eta) = f(\eta)Z(\eta) + 1 \quad (193)$$

where $Z(\eta) = \int_0^\infty e^{-\eta u}\overline{\mathbf{\Psi}}_0(u)du \quad (194)$

$$f(\eta) = \int_0^\infty f_0(u)e^{-\eta u}du = \frac{(M+1)^2}{4M}\,\frac{1 - e^{-\eta}}{1 + \eta}$$

Hence $$Z(\eta) = \frac{1}{1 - f(\eta)} \quad (195)$$

Taking the Laplace inverse,

$$\overline{\mathbf{\Psi}}_0(u) = \frac{1}{2\pi i}\int_{-i\infty}^{+i\infty} Z(\eta)e^{\eta u}d\eta \quad (196)$$

where the contour is taken to the right of all the poles (*cf.* Doetsch). If we wish to know $\overline{\mathbf{\Psi}}_0(u)$ for large u, we need only investigate $Z(\eta)$ for small η in accordance with the Tauberian theorems. For small η,

$$f(\eta) = \int_0^\infty f_0(u)\,du\left(1 - \eta u + \frac{\eta^2u^2}{2} \cdots\right) \quad (197)$$

$$\cong 1 - \eta\bar{u} = 1 - \eta\xi$$

with $$\xi = 1 - \frac{(M-1)^2}{2M}\ln\frac{M+1}{M-1}$$

Thus from Eqs. 195 and 196:

$$\overline{\mathbf{\Psi}}_0(u) \sim \frac{1}{2\pi i}\int_{-i\infty}^{+i\infty}\frac{e^{\eta u}}{\xi\eta}\,d\eta = \frac{1}{\xi} \quad (198)$$

Since the integral of the slowing-down density over all space is:

$$Q = \int_{-\infty}^{\infty} qdz = \xi\overline{\mathbf{\Psi}}_0(u) \quad (199)$$

Eq. 198 implies

$$Q \sim 1 \quad (200)$$

This is what one would obtain from Eq. 149 which was derived from age theory. The fact that age theory gives the exact asymptotic result is the principal justification for making the approximations in the manner done in Part III (*i.e.*, making use of the variables Ψ_0 and u and keeping only the first derivative of Ψ_0 with respect to u).

The Laplace inverse in Eq. 196 can easily be evaluated for u less than ϵ

by means of Eq. 195. We find:

$$\overline{\Psi}_0(u) = \alpha e^{(\alpha-1)u} \qquad u < \epsilon \quad (201)$$

with $$\alpha = \frac{(M+1)^2}{4M}$$

For hydrogen, $\epsilon = \infty$, $\alpha = 1$, $\xi = 1$, so that Eq. 201 holds in general, yielding:

$$\overline{\Psi}_0(u) = 1 \quad (202)$$

The behavior of $\overline{\Psi}_0(u)$ for intermediate values of u when $M > 1$ was investigated by Placzek (*28*). He showed that $\overline{\Psi}_0(u)$ oscillates about the asymptotic value with an amplitude that rapidly decreases as u increases. The oscillations effectively die out completely when u is two or three times ϵ. At $u = \epsilon$, $\overline{\Psi}_0(u)$ has a large discontinuity (*cf.* Fig. 1 of Placzek's paper). This discontinuity occurs because neutrons cannot go beyond $u = \epsilon$ in a single collision.

For small capture [that is, $h(u)$ close to one] Eq. 192 can be treated as above by writing:

$$h(u')\overline{\Psi}_0(u') = h(u)\overline{\Psi}_0(u) + (u' - u)\frac{\partial h(u)\overline{\Psi}_0(u)}{\partial_u} \quad (203)$$

where u is not too close to zero. Then Eq. 192 becomes:

$$\overline{\Psi}_0(u) = h(u)\overline{\Psi}_0(u) - \xi\frac{\partial(h\overline{\Psi}_0)}{\partial u} \quad (204)$$

so that [since $h(u) \cong 1$]:

$$\overline{\Psi}_0 \cong \frac{1}{\xi} e^{-\int_0^u \frac{[1-h(u')]}{\xi} du'} \quad (205)$$

which is equivalent to Eq. 191.

In the case of resonance absorption it is not generally possible to assume that $h(u)$ is close to one. However, Placzek has shown how resonance absorption may be treated if the absorption extends over a logarithmic energy interval which is only a few times ϵ. Let us assume that the absorption is zero for u less than u_0, and large in a short range beyond u_0 (*cf.* Fig. 10).

If u_0 is large compared to ϵ, then for u less than u_0 the solution of Eq. 192 is:

$$\overline{\Psi}_0(u) \cong \frac{1}{\xi} \quad (206)$$

Placzek showed that under the assumption that $\overline{\Psi}_0(u) = 1/\xi$ for $u < u_0$, the solution for u between u_0 and $u + \epsilon$ is:

$$\overline{\Psi}_0(u) = \frac{1}{\xi} e^{P(u)} \left[1 - \alpha e^{-\epsilon}\int_{u_0}^{u} e^{-P(u')}du'\right] \quad (207)$$

where: $$P(u) = \alpha\left[e^{-\epsilon}(u - u_0) - \int_{u_0}^{u} g(u')du'\right]$$

with $g = 1 - h$ and $\alpha = (M+1)^2/4M$. It is possible to extend very simply the solution to the intervals $u_0 + \epsilon$ to $u_0 + 2\epsilon$, etc. The rate at which neutrons are being absorbed in the resonance is:

$$\int_{\text{resonance}} duNv(\sigma - \sigma_s) = \int_{\text{resonance}} du\overline{\Psi}_0(u)[1 - h(u)] \quad (208)$$

Hence, the resonance may be considered as a negative source of neutrons of magnitude indicated by Eq. 208, so that for $u \gg u_0$ the solution is

$$\overline{\Psi}_0(u) = \frac{1}{\xi}\left(1 - \int_{\text{resonance}} du'\overline{\Psi}_0(u')[1 - h(u')]\right) \quad (209)$$

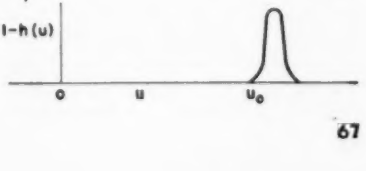
where $\overline{\Psi}_0(u')$ is given by Eq. 201, or the appropriate extension if the resonance is broader than ϵ.

If the resonance has logarithmic width small compared to ϵ, the foregoing method yields a simple result. Then Eq. 207 reduces to (since $\alpha\Delta u \ll 1$):

$$\overline{\Psi}_0(u) \cong \frac{1}{\xi} \quad (210)$$

so that from Eq. 203:

FIG. 10



$$\overline{\Psi}_0(u) = \frac{1}{\xi}$$

$$\left(1 - \int_{\text{resonance}} du' \frac{[1 - h(u')]}{\xi}\right) \quad (211)$$

or $\Delta \ln \overline{\Psi}_0 =$

$$-\frac{1}{\xi}\int_{\text{resonance}} du'[1 - h(u')] \quad (212)$$

since under our assumption the fractional change in $\overline{\Psi}_0$ is small.

If the absorption is small except in very narrow resonances which are separated by logarithmic energy intervals large compared to ϵ, then Eqs. 205 and 212 may be combined, giving:

$$Q(u) = \xi\overline{\Psi}_0(u) =$$

$$\exp\left(-\int_0^u \frac{\sigma_a}{\sigma_a + \sigma_s}\frac{du'}{\xi}\right) \quad (213)$$

where σ_a and σ_s are absorption and scattering cross sections, respectively.

We have used the relation

$$1 - h(u) = 1 - \frac{\sigma_s}{\sigma} = \frac{\sigma_a}{\sigma_a + \sigma_s} \quad (214)$$

Equation 213 is not valid for u slightly greater than 0, or slightly greater than the value corresponding to a resonance.

Note that the number of neutrons absorbed by a narrow resonance does not approach infinity as σ_a approaches infinity since the ratio $\sigma_a/(\sigma_a + \sigma_s)$ remains finite. This arises physically because neutrons can jump across the resonance energy in a collision so that the greatest effect that the resonance can have is to absorb all the neutrons which happen to land in the region where σ_a is large.

BIBLIOGRAPHY

Bibliography items referred to in the preceding text can be found in the complete bibliography published with Part I of this series of papers; see NUCLEONICS **4**, No. 5, 22 (1949).

THERE ARE MANY EXAMPLES of discoveries basic to the present work of the Atomic Energy Commission which were in fact made by Communists or Communist sympathizers. Of these many examples, we may cite a famous one: The major—one might almost say the only—present peaceful application of atomic energy rests on the preparation and use of artificial radioactive materials, which were discovered by Joliot, who is a Communist, and by his wife, who is a Communist sympathizer. It would be folly to suppose that the United States would be the stronger, or our science and industry the more vigorous, if this discovery had not been made. It would be contrary to all experience to suppose that only those who throughout their lives have held conformist political views would make the great discoveries in the future. The people and the Government of the United States have a stake in scientific discovery and invention; and it is for this stake, rather than as an act of benevolence toward the recipients of the grants-in-aid, that one must look for justification for having a fellowship program at all.

—J. Robert Oppenheimer, from a letter to the chairman of the Joint Congressional Atomic Energy Committee, introduced at the Committee hearings on the AEC fellowship program on May 17

BOOKS

DIE STATISTISCHE THEORIE DES ATOMS UND IHRE ANWENDUNGEN, (The Statistical Theory of the Atom and its Applications) by Professor Dr. P. Gombás, Springer, Vienna, 1949, 406 pages, $18.60. *Reviewed by S. A. Goudsmit, Research Physicist, Brookhaven National Laboratory, Upton, L. I.*

This book by Professor Gombás of Budapest, who is now in the United States, covers the complete development of the statistical treatment of the atom from its foundations by Thomas and Fermi in 1926, to its applications up to about 1940. Some references to later papers, almost exclusively Hungarian, are included, but it seems that the statistical atom was considered essentially a finished chapter of physics in the late thirties.

The first 165 pages form an excellent textbook on the statistical model. This section can be highly recommended to students and to all who need a working knowledge of the statistical atom. It includes treatments of the exchange and relativistic corrections, perturbations and the hydrodynamic model of Bloch.

The second part of the book covers the applications. There is first a chapter on special atomic problems, such as, the statistical computation of excitation energies, stopping power of atoms, scattering of X-rays and electrons and several other problems in which the statistical model plays a role.

The following chapters deal with applications to molecules, crystals, the theory of metals and matter under high pressure. It must be stated, however, that this part of the book is more a compilation or reference work than a textbook. Only such details as are directly connected with the statistical atom are discussed fully. These last 230 pages could probably have been filled more advantageously if the author had made a critical selection of the multitude of material available and treated it as he did the first part of his book.

There is one strange and conspicuous omission, no mention is made of the statistical models of the atomic nucleus. A score of pages on this subject might have transformed this book from a treatise for specialists into a best-seller among modern nuclear scientists.

FOUNDATIONS OF NUCLEAR PHYSICS, with a bibliography compiled by Robert T. Beyer, Dover Publications, Inc., New York, 1949, 272 pages, $2.95.

This book contains facsimiles of thirteen fundamental studies as they were originally reported in the scientific journals. The following are the titles of the papers included: The Positive Electron by Carl D. Anderson; The Existence of a Neutron by J. Chadwick; Experiments with High-Velocity Positive Ions by J. D. Cockroft and E. T. S. Walton; Physique Nucléaire—Un Nouveau Type de Radioactivité by I. Curie and M. F. Joliot; Possible Production of Elements of Atomic Number Higher than 92 by E. Fermi; Versuch einer Theorie der β-Strahlen by E. Fermi; Über die Magnetische Ablenkung von Wasserstoffmolekülen und das Magnetische Moment des Protons by R. Frisch and O. Stern; Zur Quantentheorie des Atomkernes by G. Gamow; Über den Nachweis und das Verhalten der bei der Bestrahlung des Urans Mittels Neutronen Entstehenden Erdalkalimetalle by O. Hahn and F. Strassmann; The Production of High Speed Light Ions without the Use of High Voltages by E. O. Lawrence and M. S. Livingston; The Scattering of α and β Particles by Matter and the Structure of the Atom by E. Ruther-

ford; Collision of α Particles with Light Atoms, Part IV—An Anomalous Effect in Nitrogen by E. Rutherford; On the Interaction of Elementary Particles by H. Yukawa.

TABLES OF SINES AND COSINES TO FIFTEEN DECIMAL PLACES AT HUNDREDTHS OF A DEGREE, Applied Mathematics Series No. 5, National Bureau of Standards, Washington, D. C., 1949, 95 pages, $0.40. *Reviewed by Philip H. Lowry, Brookhaven National Laboratory, Upton, L. I.*

A convenient arrangement of sines and cosines in parallel columns with the second central differences. A supplementary table simplifies interpolation by Everett's formula to 15-place accuracy for ten-thousandths of a degree. The first three coefficients of Taylor's expansion are given to 16 places, allowing full accuracy in interpolation by that method. Herrmann's table of sines and cosines to thirty decimal places at intervals of one degree is reprinted.

This table fills a long-felt want. The only comparable table was published in 1633 and is practically unobtainable.

Copies are obtainable from the Supt. of Documents, Washington 25, D. C. Remittances from foreign countries must be made in U. S. exchange and must include an additional sum of one third the publication price to cover mailing costs.

ISOTOPIC REPORT, Tabular Survey of the Properties of Atomic Nuclei as Known at the End of 1948, by J. Mattauch and A. Flammersfeld, in German and English. This is a special issue of the *Zeitschrift für Naturforschung*, 1949, 244 pages.

Data published since 1942 have been tabulated in this report, according to a press release from the publisher. It contains details on about 300 stable and 700 unstable nuclei, 2,000 different nuclear reactions, and a bibliography of more than 2,000 papers. The Plutonium Project Report and the Table of Isotopes by Seaborg and Perlman [*Revs. Mod. Phys.* **20,** 585–667 (1948)] are incorporated. This report will be followed by supplements every two or three years.

Orders should be addressed to Verlag der *Zeitschrift für Naturforschung*, Tübingen, Johannesweg 11, Germany, French zone.

BOOKS RECEIVED

INTRODUCTION TO RADIOCHEMISTRY, by G. Friedlander and J. W. Kennedy, John Wiley and Sons, Inc., New York, 1949, 412 pages, $5.00.

THE PHARMACOLOGY AND TOXICOLOGY OF URANIUM COMPOUNDS, edited by Carl Voegtlin and Harold C. Hodge, lithographed by Edwards Bros., Ann Arbor, Mich., published by McGraw-Hill Book Co., Inc., New York, 1949, 1084 pages, 2 parts, $10.

RADIOACTIVE TRACER TECHNIQUES, by G. K. Schweitzer and Ira B. Whitney, D. Van Nostrand Co., Inc., New York, 1949, 241 pages, $3.25.

ATOMIC ENERGY YEARBOOK, edited by John Tutin, Prentice-Hall, Inc., New York, 1949, 237 pages, $3.85.

NUCLEONIC EVENTS

PROGRAM ANNOUNCED FOR IRE-AIEE CONFERENCE

The second annual "Conference on Electronic Instrumentation in Nucleonics and Medicine," jointly sponsored by the Institute of Radio Engineers and the American Institute of Electrical Engineers, will follow a pattern similar to the one held last year. To take place at the Hotel Commodore in New York on October 31, and November 1–2, the meeting will be devoted to medical electronics on the first day, nucleonics in medicine on the second day, and the physical aspects of nucleonics instrumentation on the third day.

On the evening of November 1, a round-table discussion on "Evaluation of Radiation Hazards" will be held at the Hotel Commodore. A four-man panel of scientific experts will lead the discussion and will answer questions from the audience. Following this, Karl T. Compton, chairman of the Research and Development Board of the National Military Establishment, will deliver an address.

Listed below are those papers for which speakers have already accepted:

October 31

Audible Interpretation of Electroencephalograph—Robert R. Riesz, Bell Telephone Laboratories, Murray Hill, N. J.

High Fidelity Electrocardiography—S. R. Gilford, National Bureau of Standards, Washington, D. C.

Electrical Methods of Blood-pressure Recording—Frank Noble, College of Electrical Engineering, Cornell University, Ithaca, N. Y. Discussion by William C. Ballard, College of Electrical Engineering Cornell University, Ithaca, N. Y.

Stable D-C Amplifier for Biological Recordings—Harry Grundfest, College of Physicians and Surgeons, Columbia University, N. Y.

Design of Cathode-Ray Oscillography for Biological Applications—W. A. Geohegan, Cornell Medical College, New York, N. Y.

November 1

Medical Applications of Ionizing Radiation—Edith Quimby, Radiological Research Laboratory, Columbia University, N. Y.

Dosage Measurements of Ionizing Radiation—G. Failla, Radiological Research Laboratory, Columbia University, N. Y.

Measurement of Low-Energy Beta-Ray Emitters: Ionization Chambers—Norman Bailey, Radiological Research Laboratory, Columbia University, N. Y.; **Proportional Counters**—C. J. Borkowski, Oak Ridge National Laboratory, Oak Ridge, Tenn.; **Gas-Filled Counters for C^{14} and H^3**—M. L. Eidinoff, Sloan-Kettering Institute, New York, N. Y.

November 2

An Automatic Isodose Recorder—G. J. Hine, Sloan-Kettering Institute, New York, N. Y.

Energy Transfer in Scintillation Counting Systems—G. A. Morton, RCA Research Laboratories, Princeton, N. J.

Phosphors for Scintillation Counters—R. H. Gillette, Linde Air Products Laboratories, Tonawanda, N. Y.

Solids for Radiation Detection—R. M. Lichtenstein, General Electric Company, Schenectady, N. Y.

Some Design Features of Electrical Counting Systems—N. F. Moody, National Research Council of Canada, Chalk River, Ontario.

Desirable Improvements in Nuclear Instruments—J. B. H. Kuper, Brookhaven National Laboratory, Upton, L. I.

Criteria in the Selection of Radioisotopes for Industrial Use—Eric Clark, Tracerlab, Inc., Boston, Mass.

HARVARD DEDICATES NUCLEAR LABORATORY

The new nuclear studies laboratory at Harvard University was formally opened June 15. At the heart of the laboratory is a 125-Mev cyclotron, said to be the only one operating or planned for the 30–200 Mev range.

The machine was financed by a $1,000,000 grant by the Office of Naval Research and the Atomic Energy Commission. Harvard spent $450,000 for the building.

U. OF CHICAGO BUILDS NEW RESEARCH LAB

The cornerstone of the University of Chicago's $7,500,000 laboratory for nuclear, biological and metallurgical research was laid on June 21 by Enrico Fermi, Nobel-prize winner and distinguished service professor of physics at the university. This four-story structure is the second unit of the $12,000,000 research center comprising the Institute for Nuclear Studies, the Institute for the Study of Metals, and the Institute of Radiobiology and Biophysics.

The first unit, the accelerator building (a $1,250,000 one-story laboratory and basement), houses the university's new 170-inch synchrocyclotron, which is scheduled for operation in early 1950. One of the three largest cyclotrons in the world, the $2,200,000 instrument was financed by the Office of Naval Research and the public contributions to the university's Cancer Research Foundation.

A 100-Mev betatron for research on high-energy gamma rays is also located in the accelerator building.

AT & T SUBSIDIARIES TO OPERATE SANDIA BASE

The Western Electric Company and the Bell Telephone Laboratories, subsidiaries of the American Telephone and Telegraph Company, have replaced the University of California as operator of the Sandia Laboratory at Sandia Base, Albuquerque, N. M. Sandia is primarily an atomic weapon engineering center.

The Sandia Laboratory has been operated since 1945 by the University of California under its contract for the operation of the Los Alamos Scientific Laboratory. Sandia has grown from a small liaison group representing Los Alamos into a major facility. The University of California advised the commission last winter that the university felt it should not continue to manage Sandia as a part of the scientific research program of the Los Alamos Laboratory.

Operations of both Los Alamos and Sandia are carried out under the AEC's Santa Fe office, of which Carroll L. Tyler is manager. Paul J. Larsen is director of the Sandia branch.

AEC MAKES SURVEY OF TRINITY SITE

A small crew of University of California scientists under contract with the Atomic Energy Commission are continuing during the summer to survey Trinity, the site of the world's first atomic bomb explosion, which took place near Alamogordo on the morning of July 16, 1945.

Similar surveys of the Trinity site and environs have been made over the past

two summers to study the fate of radioactive materials deposited in or on a desert soil. Many problems of the redistribution of radioactive materials in soil, plant and animal life cycles are under study.

Four years ago the test bomb blast in this desert area denuded the soil of vegetation in an area several hundreds of feet in radius. This area now has been reinvaded by woods and grasses and it is said that it will, in due time, become reestablished with the typical desert vegetation.

Field parties have studied mice, rats, rabbits, snakes, lizards and birds captured in the immediate area of the crater and in the area contaminated by "fall-out" from the detonation. ("Fall-out" refers to radioactive bomb particles and dust which gradually settled out of the atmosphere after the explosion.) These animals are reported to be normal and in good health. No significant amounts of radioactivity were found in the animal tissues.

In particular researchers looked for abnormal specimens and for observable effects on cattle said to have grazed over contaminated areas for two or three years. No such effects were discovered.

AUSTRALIA RECEIVES ISOTOPES FROM ENGLAND

The first shipment of radioisotopes from England to Australia since the British reactors started operation was received at Sydney last month. This first shipment consisted of radiophosphorus for distribution by the Commonwealth X-ray and Radium Laboratory for use in medical treatment.

Over the past two years various types of isotopes have been procured from the U. S. Atomic Energy Commission. With the recent commencement of the production of radioisotopes at the British Atomic Energy Research Establishment at Harwell, Australia has found that by making purchases from Britain it can conserve dollars. Thus, these trial shipments have started.

KELLEX CORP. GETS NEW LAB FOR CHEMICAL STUDIES

New laboratory facilities for chemical and physical research by the Kellex Corporation for the Atomic Energy Commission are now in operation at the Jersey City, N. J., plant of the M. W. Kellogg Company, parent company of the Kellex Corporation. Constructed under a prime contract between the Kellex Corporation and the AEC, the research facilities will enable Kellex scientists and engineers to develop and test on a laboratory scale certain chemical processes concerned with the commission's nuclear reactor development program.

The Kellex studies form an integral part of the commission's research, development and construction program aimed at the improvement of chemical separation of plutonium from uranium. Through this program it is said that more efficient use of the available material will be realized by improved separation and recovery processes with resulting great economies in manufacture.

One of the features of the Kellex laboratory set-up is a test floor adjacent to the laboratory proper, which is completely equipped with utility services and other necessary facilities. Here component equipment and techniques developed for the project will be tested and proved.

The laboratory will be under the leadership of W. A. Bain, whose promotion to director of chemical research of Kellex was recently announced.

U. OF ARKANSAS TO STUDY "HOT-ATOM" CHEMISTRY

The Institute of Science and Technology of the University of Arkansas has received a $25,000 contract from the Atomic Energy Commission to

conduct research on the chemical effects of nuclear transformations. An effort will be made to utilize "hot-atom" exchange reactions in the preparation of tracer compounds, particularly those useful in medical and biological research. The project will also include a study of the chemical effects of beta decay processes.

Raymond R. Edwards, associate chemist in charge of nuclear studies at the institute, will head the project. Paul L. Day and John R. Totter, of the School of Medicine's department of biochemistry, will be collaborators.

WING OF HARWELL "HOT" LAB COMPLETED

One wing of the new radiochemical laboratory at the Atomic Energy Research Establishment, Harwell, England, has been completed. The laboratory will be used primarily for research on the chemical problems of separating the fissile element plutonium from uranium rods that have been irradiated in the pile. The laboratory also will be used for other chemical work with radioactive materials, including the separation of radioisotopes produced in BEPO (British Experimental Pile), and processing materials that have been irradiated in the pile.

U. OF MICHIGAN TO STUDY ANCIENT PLANT, ANIMAL LIFE

The University of Michigan's atomic energy research group has made an initial grant of $1,500 to start research on a method of dating the remains of animal and plant life of the last 30,000 years with a degree of accuracy which was not possible until recently. This method for dating was developed by W. F. Libby and his associates at the University of Chicago's Institute for Nuclear Studies.

This initial grant (made by the preliminary planning committee of the Phoenix Project) will be spent under the direction of H. R. Crane, professor of physics, in the construction and testing of the necessary instruments. Once Dr. Crane has constructed the necessary instruments, responsibility of collecting specimens for dating will fall to a committee headed by James B. Griffin, associate professor of anthropology and director of the Museum of Anthropology.

'49 INSTRUMENT CONVENTION TO BE HELD IN ST. LOUIS

The Instrument Society of America will hold its 1949 conference and exhibit at the St. Louis Auditorium, September 12–16. A technical program of 75 papers in 26 sessions has been arranged by the co-sponsoring societies.

The Instrument Society of America will have three sessions on instrumentation for production processes; two on inspection and gaging; two on maintenance and operation of instruments; one on instruments in transportation; three on instruments in testing; and two on instruments in chemical analysis. The National Telemetering Forum will have four sessions on guided missile instrumentation. The American Institute of Physics will have three sessions on scientific instruments. The American Society of Mechanical Engineers, Industrial Instruments and Regulators Division, will have three sessions. The American Institute of Electrical Engineers, Instruments and Measurements Committee, will have two sessions.

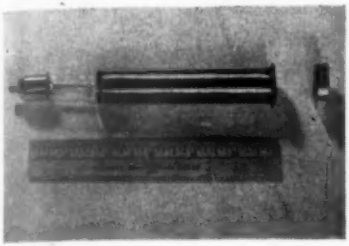
AMA MERGES TWO JOURNALS

On January 1, the *Journal of Industrial Hygiene* and *Occupational Medicine* will be merged to form *Archives of Industrial Hygiene and Occupational Medicine*. The new journal, published by the American Medical Association, will have as its editor Philip Drinker of the Harvard School of Public Health.

PRODUCTS and MATERIALS

GEIGER TUBES

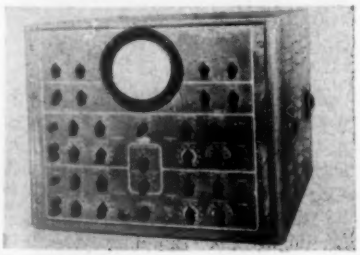
Electronic Products Co., 111 E. 3rd St., Mt. Vernon, N. Y. This company has



a new line of radiation detection tubes. Among them is a series of selfquenching, high sensitivity, beta-gamma tubes. These feature a new filling gas with halogen admixture and a new processing technique which is said to make the tubes immune to the effects of temperature changes and to excess voltages.

4-BEAM C.R.O.

Allen B. DuMont Laboratories, Inc., 1000 Main Ave., Clifton, N. J. The Four-Beam Cathode-Ray Indicator can display simultaneously four related or unrelated, independent phenomena on a single cathode-ray tube screen. The instrument contains its own power supply and a horizontal amplifier for each of the four channels. Sweep circuits



and vertical amplifiers are not provided because the instrument is intended for use with an oscillograph-record camera of the moving-film type. The motion of the film thus provides the time base.

LIGHTWEIGHT COUNTER

Nuclear Instrument and Chemical Corp., 223 W. Erie St., Chicago 10, Ill. This Geiger counter "Sniffer" has been designed specifically for the vacationer

who may be interested in prospecting for radioactive ores. Weighing only 2 lbs, the counter is the size of a folding camera. It uses two ordinary flashlight batteries from which, it is claimed, 1,200 volts can be obtained. Presence of radioactive materials is indicated by clicking in a pair of headphones.

COUNTING-RATE METER

Kelley-Koett Mfg. Co., 212 W. 4th St., Covington, Ky. The model K-800 beta-gamma counting-rate meter uses a GM tube probe which has a movable shield for differentiating between beta and gamma radiation. The shield is 2 mm stainless steel which is said to exclude all but the very highest energy

beta rays. A scale selector switch permits choice of three gamma ranges—0.2, 2.0 and 20.0 mr/hr. The scale is also calibrated in 360, 3,600 and 36,000 counts per min. An earphone is provided for aural monitoring. The meter weighs 9 lbs and its probe can be extended to about 30 in.

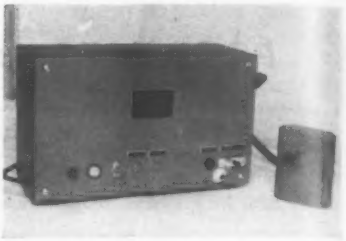
GAMMA GEIGER TUBE

Sylvania Electric Products, Inc., Electronics Div., 500 Fifth Ave., New York 18, N. Y. The GG306 gamma-ray detector tube, which is 6 in. long and 2 in. in diameter, is selfquenching;

operates at 960 volts; has an average Geiger threshold of 900 volts; provides 810 counts per minute from 5 micrograms of radium filtered through ⅛ in. of lead 12 in. from end; has a maximum recovery time of 800 microseconds; a maximum dead time of 100 microseconds; is suitable for an input circuit impedance of 5 megohms; and has an ambient temperature rating of 40° C to −70° C.

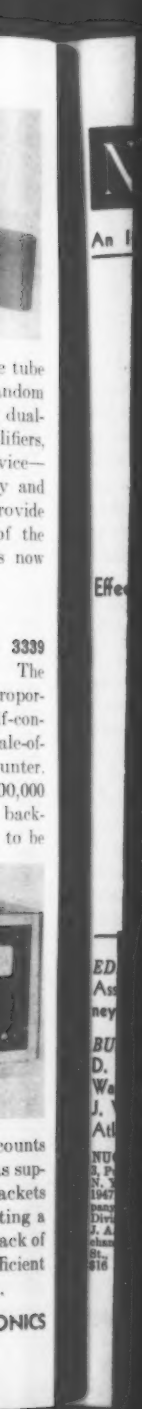
COINCIDENCE AMPLIFIER

W. S. Macdonald Co., Inc., 33 University Rd., Cambridge 38, Mass. The type 152A coincidence amplifier is designed for use with scintillation counters and Geiger-Müller tubes. The use of coincidence in radiation research increases the sensitivity by blocking noise signals and thus allowing higher amplifier gain. Desired signals which



simultaneously appear at the gate tube are permitted to pass, while random noise is rejected. This unit is a dual-channel device with suitable amplifiers, limiting circuits and gating device—self-contained with power supply and controls. It is designed to provide proper output to operate any of the known scalers or pulse counters now on the market.

PROPORTIONAL COUNTER

Nuclear Measurements Corp., 3339 Central Ave., Indianapolis 5, Ind. The model PC-1 alpha-beta-gamma proportional counter is a completely self-contained unit. It comprises a scale-of-512 with a resettable six-digit counter. This combination counts up to 500,000 counts per minute. The alpha background of the instrument is said to be

easily maintained at less than 5 counts per hr. The accessory built-in gas supply comprises a regulator and brackets with hold-down straps for mounting a 4 in. × 12 in. tank of gas on the back of the cabinet. This supply is sufficient for roughly 2,000 sampling cycles.